Practical error bounds for properties in plane-wave electronic structure calculations

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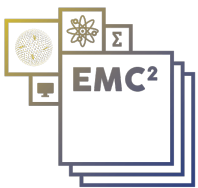

Quantum mechanics of noninteracting electrons

We consider the stationary Schrödinger equation

$$\begin{cases} H_0\varphi_i = \varepsilon_i\varphi_i, \ \varepsilon_1 \leqslant \cdots \leqslant \varepsilon_N, \\ \|\varphi_i\|_{L^2} = 1, \end{cases} \qquad H_0 := -\frac{1}{2}\Delta + V$$

where φ_i is the wavefunction associated to electron i. Then,

- $E = \sum_{i=1}^{N} \varepsilon_i$ is the total energy;
- $\rho(x) = \sum_{i=1}^{N} |\varphi_i(x)|^2$ is the total electronic density.

Numerical resolution

$$\text{Find } \varphi_i \in \mathbb{C}^{\mathcal{N}}, \text{ s.t } H_0\varphi_i = \varepsilon_i\varphi_i, \quad \varepsilon_1 \leqslant \cdots \leqslant \varepsilon_N$$

Orbitals φ_i are not unique (degeneracies, phase factor) $\rightsquigarrow$ better to work with the *projectors* onto the space spanned by the $(\varphi_i)_{1\leqslant i\leqslant N}$:

$$P := \sum_{i=1}^{N} |\varphi_i\rangle \langle\varphi_i| \in \mathbb{C}^{\mathcal{N}\times\mathcal{N}}_{\text{herm}}.$$

- P is a rank N orthogonal projector (*density matrices*);
- the total energy then writes

$$E = \sum_{i=1}^{N} \varepsilon_i = \sum_{i=1}^{N} \langle\varphi_i|H_0\varphi_i\rangle = \mathrm{Tr}(H_0P),$$

and is minimal for this P among all rank N orthogonal projectors.

We have two equivalent problems:

$$\begin{cases} H_0\varphi_i = \varepsilon_i\varphi_i, \ \varepsilon_1 \leqslant \cdots \leqslant \varepsilon_N, \\ \|\varphi_i\|_{L^2} = 1, \end{cases} \quad \Leftrightarrow \quad \min_{P\in\mathcal{M}_N} \mathrm{Tr}(H_0 P)$$

where

$$\mathcal{M}_N := \left\{ P \in \mathbb{C}^{\mathcal{N}\times\mathcal{N}} \;\middle|\; P = P^*, \ \mathrm{Tr}(P) = N, \ P^2 = P \right\}$$

is the set of rank N orthogonal projectors. It is a *Grassmann* manifold.

General framework

In reality, electrons *do* interact together so that the general form of the energy is

$$E(P) \coloneqq \mathrm{Tr}\,(H_0 P) + E_{\mathrm{nl}}(P),$$

where

- $P \in \mathbb{C}^{\mathcal{N}\times\mathcal{N}}_{\mathrm{herm}}$ is a density matrix;
- H_0 is the core Hamiltonian;
- E_{nl} models the electron-electron interaction depending on the model (Kohn-Sham DFT, Hartree-Fock, Gross-Pitaevskii, ...).

$$\min_{P\in\mathcal{M}_N} E(P) = \mathrm{Tr}\,(H_0 P) + E_{\mathrm{nl}}(P),$$

$$\mathcal{M}_N \coloneqq \left\{ P \in \mathbb{C}^{\mathcal{N}\times\mathcal{N}} \;\middle|\; P = P^*,\ \mathrm{Tr}(P) = N,\ P^2 = P \right\}.$$

Let $\mathcal{H} \coloneqq \left(\mathbb{C}^{\mathcal{N}\times\mathcal{N}}_{\mathrm{herm}}, \|\cdot\|_{\mathrm{F}}\right)$, endowed with the Frobenius scalar product $\mathrm{Tr}(A^*B)$.

Assumption 1 $E_{\mathrm{nl}} : \mathcal{H} \to \mathbb{R}$ is twice continuously differentiable, and thus so is E.

Assumption 2 $P_* \in \mathcal{M}_N$ is a nondegenerate local minimizer in the sense that there exists some $\eta > 0$ such that, for $P \in \mathcal{M}_N$ in a neighborhood of P_*, we have

$$E(P) \geqslant E(P_*) + \eta \|P - P_*\|_{\mathrm{F}}^2.$$

In practice, the required $\mathcal{N}$ to achieve high precision is way too high. To solve this issue, we use subspaces of smaller dimension to compute a variational approximation of P_*, the reference solution in $\mathcal{M}_N$.

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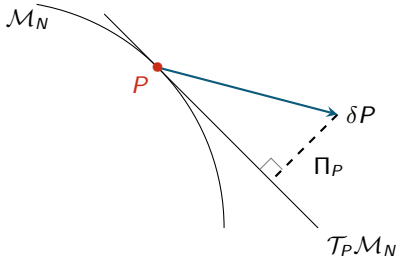
Question : How to evaluate the error made on quantities of interest (QoI) ? We focus here on the energy, the density or the forces.

Structure of the manifold: the tangent space

$\mathcal{M}_N$ is a smooth manifold, we can define its tangent space (it is a $\mathbb{R}$ vector space). Π_P is the orthogonal projection on $\mathcal{T}_P\mathcal{M}_N$:



A density matrix $P \in \mathcal{M}_N$ can be described with N orbitals (any orthonormal basis of $\mathrm{Ran}(P)$):

$$P = \sum_{i=1}^{N} |\varphi_i\rangle \langle\varphi_i| \quad \text{with} \quad \langle\varphi_i|\varphi_j\rangle = \delta_{ij}.$$

Given such a P, an element X of $\mathcal{T}_P\mathcal{M}_N$ can be described with N vectors that are all orthogonal to the φ_i's:

$$X = \sum_{i=1}^{N} |\varphi_i\rangle \langle\psi_i| + |\psi_i\rangle \langle\varphi_i| \quad \text{with} \quad \langle\varphi_i|\psi_j\rangle = 0,$$

$$\Rightarrow \|X\|_{\mathrm{F}}^2 = 2 \sum_{i=1}^{N} \|\psi_i\|^2$$

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$$\begin{array}{rcl} P \in \mathcal{M}_N & \leftrightarrow & (\varphi_i)_{1\leqslant i\leqslant N} \in (\mathbb{C}^{\mathcal{N}})^N \text{ spanning } \mathrm{Ran}(P) \\ X \in \mathcal{T}_P\mathcal{M}_N & \leftrightarrow & (\psi_i)_{1\leqslant i\leqslant N} \in (\mathbb{C}^{\mathcal{N}})^N \text{ where } \langle\varphi_i|\psi_j\rangle = 0 \end{array}$$

Change of norm : given $X \in \mathcal{T}_P\mathcal{M}_N$, one might want to compute $\|\boldsymbol{M}X\|_{\mathrm{F}}$ for a metric $\boldsymbol{M}$ on the tangent space. This can be translated in terms of orbitals as

$$\boldsymbol{M}X = \sum_{i=1}^{N} |\varphi_i\rangle \langle M_i\psi_i| + |M_i\psi_i\rangle \langle\varphi_i|, \quad \|\boldsymbol{M}X\|_{\mathrm{F}} = 2\sum_{i=1}^{N} \|M_i\psi_i\|$$

where $M_i : \mathrm{Ran}(\{\varphi_j\})^\perp \to \mathrm{Ran}(\{\varphi_j\})^\perp$ and can eventually depend on the band i. In this talk we will use (with Π the projection on $\mathrm{Ran}(\{\varphi_j\})^\perp$ and t_i the kinetic energy of band i):

$$\begin{array}{rcll}
\boldsymbol{M}^{1/2} & \leftrightarrow & \Pi(t_i - \Delta/2)^{1/2}\Pi & \leftrightarrow \quad \mathrm{H}^{1/2} \text{ norm} \\
\boldsymbol{M} & \leftrightarrow & \Pi(t_i - \Delta/2)^{1/2}\Pi(t_i - \Delta/2)^{1/2}\Pi & \leftrightarrow \quad \mathrm{H}^{1} \text{ norm} \\
\\
\boldsymbol{M}^{-1/2} & \leftrightarrow & (\Pi(t_i - \Delta/2)^{1/2}\Pi)^{-1} & \leftrightarrow \quad \mathrm{H}^{-1/2} \text{ norm} \\
\boldsymbol{M}^{-1} & \leftrightarrow & (\Pi(t_i - \Delta/2)^{1/2}\Pi(t_i - \Delta/2)^{1/2}\Pi)^{-1} & \leftrightarrow \quad \mathrm{H}^{-1} \text{ norm}
\end{array}$$

First order condition

$$\min_{P \in \mathcal{M}_N} E(P) = \mathrm{Tr}\,(H_0 P) + E_{\mathrm{nl}}(P)$$

The first-order optimality condition is $\Pi_{P_*}(H_*) = 0$, which gives

$$\boxed{P_* H_* (1 - P_*) = (1 - P_*) H_* P_* = 0},$$

where $H_* \coloneqq \nabla E(P_*)$.

In particular, $[H_*, P_*] = 0$.

Second order condition

$$\min_{P \in \mathcal{M}_N} E(P) = \mathrm{Tr}\,(H_0 P) + E_{\mathrm{nl}}(P)$$

The second order optimality condition reads

$$\boxed{\forall\, X \in \mathcal{T}_{P_*}\mathcal{M}_N,\ \langle X, (\boldsymbol{\Omega}_* + \boldsymbol{K}_*)X\rangle_{\mathrm{F}} \geqslant \eta\, \|X\|_{\mathrm{F}}^2}.$$

- $\boldsymbol{K}_* \coloneqq \Pi_{P_*} \nabla^2 E(P_*) \Pi_{P_*}$;
- the operator $\boldsymbol{\Omega}_* : \mathcal{T}_{P_*}\mathcal{M}_N \to \mathcal{T}_{P_*}\mathcal{M}_N$ is defined by,

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$\rightsquigarrow$ $\boldsymbol{\Omega}_* + \boldsymbol{K}_*$ can be interpreted as the Hessian of the energy on the manifold, $\boldsymbol{\Omega}_*$ represents the influence of the curvature.

Plane-wave DFT

Throughout the talk, we perform numerical tests in DFTK[1], a PW DFT tool-kit for Julia. In short:

- we consider a periodic system with lattice $\mathcal{R}$, ω is the unit cell and $\mathcal{R}^*$ the reciprocal lattice;
- we solve a variational approximation of the KS-DFT equations in the finite dimensional space

$$\mathcal{X}_{E_{\text{cut}}} \coloneqq \left\{ e_{\boldsymbol{G}},\ \boldsymbol{G} \in \mathcal{R}^* \;\middle|\; \frac{1}{2}|\boldsymbol{G}|^2 \leqslant E_{\text{cut}} \right\},$$

where, for $\boldsymbol{G} \in \mathcal{R}^*$,

$$\forall\, \boldsymbol{r} \in \mathbb{R}^3, \quad e_{\boldsymbol{G}}(\boldsymbol{r}) \coloneqq \frac{1}{\sqrt{|\omega|}} \exp\left(\mathrm{i}\boldsymbol{G} \cdot \boldsymbol{r}\right).$$

[1] `https://dftk.org`, developed by M. F. Herbst and A. Levitt.

Numerical setting

- FCC phase of the silicon crystal, within LDA approximation and $2 \times 2 \times 2$ Brillouin zone discretization;
- we compute a reference solution for $E_{\text{cut,ref}} = 125$ Ha $\Rightarrow$ $E_{\text{cut,ref}}$ defines $\mathcal{N}$ the size of the reference space and we obtain the reference orbitals Φ_*, the energy E_*, density ρ_*, the forces F_* on each atoms, etc...
- for smaller E_{cut}'s, we compute the associated variational approximation and we measure the error on different quantities:

$$|E - E_*|\,, \quad \|\rho - \rho_*\|_{L^2}\,, \quad |F - F_*|$$

Linearization

$\boldsymbol{\Omega}_* + \boldsymbol{K}_*$ is the Jacobian[2] of $P \mapsto \Pi_P H(P) = [P, [P, H(P)]]$ at P_*.

Thus, at first order in $\|P - P_*\|_{\mathrm{F}}^2$,

$$[P, [P, H(P)]] = [P_*, [P_*, H(P_*)]] + (\boldsymbol{\Omega}_* + \boldsymbol{K}_*)(P - P_*).$$

As $[P_*, [P_*, H(P_*)]] = 0$, with $R(P) \coloneqq [P, [P, H(P)]]$ the residual,

$$\boxed{\Pi_P(P - P_*) = (\boldsymbol{\Omega}_* + \boldsymbol{K}_*)^{-1} R(P)}$$

[2] Eric Cancès, Gaspard Kemlin, Antoine Levitt. Convergence analysis of direct minimization and self-consistent iterations. SIAM Journal of Matrix Analysis and Applications, 42(1):243–274 (2021).

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Newton's algorithm : extend the definition of $\boldsymbol{\Omega}$ and $\boldsymbol{K}$ outside of P_* and let $\mathfrak{R}$ be a retraction to the manifold

$$\boxed{P^{k+1} = \mathfrak{R}_{P^k}\left(P^k + \left(\boldsymbol{\Omega}(P^k) + \boldsymbol{K}(P^k)\right)^{-1} R(P^k)\right)}$$

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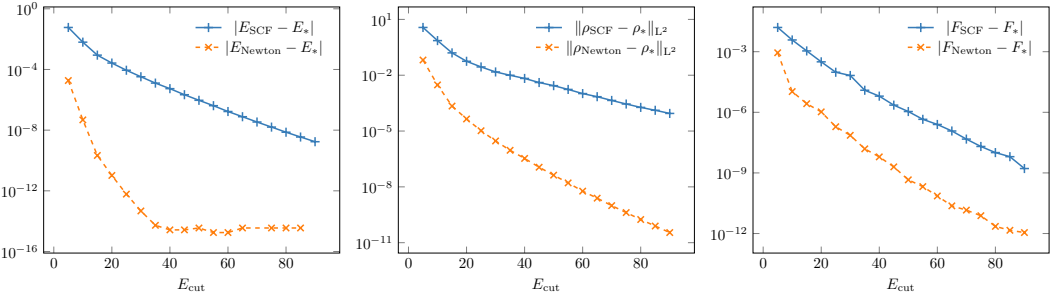
Compare DFTK QoI for given $E_{\text{cut}} < E_{\text{cut,ref}}$ and the QoI after one Newton step in the reference grid.



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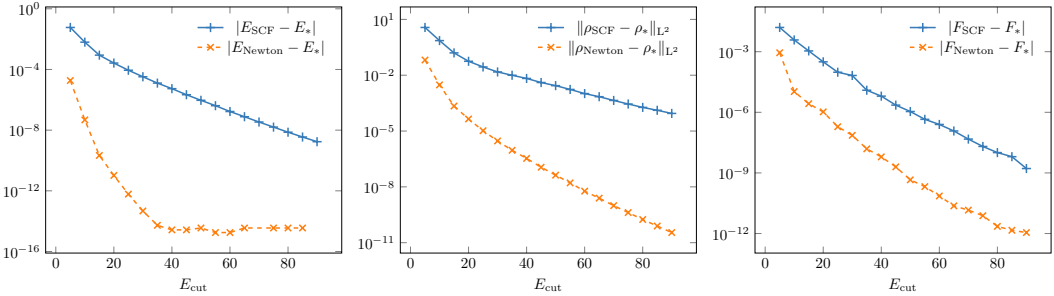

$\rightsquigarrow$ the asymptotic regime is quickly established

Error bounds based on operator norms

$$\Pi_P(P - P_*) = (\boldsymbol{\Omega}_* + \boldsymbol{K}_*)^{-1} R(P)$$

First crude bound : $\|P - P_*\|_{\mathrm{F}}$ and $\|R(P)\|_{\mathrm{F}}$ cannot be directly compared (not the same unit) but we have

$$\begin{aligned} \|P - P_*\|_{\mathrm{F}} &\approx \|\Pi_P(P - P_*)\|_{\mathrm{F}} \\ &\leqslant \left\|(\boldsymbol{\Omega}_* + \boldsymbol{K}_*)^{-1}\right\|_{\mathrm{op}} \|R(P)\|_{\mathrm{F}}\,. \end{aligned}$$

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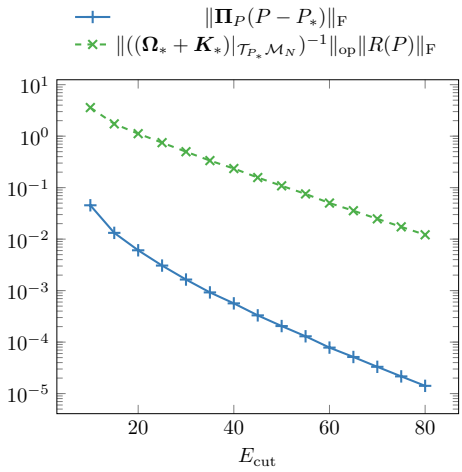
$\rightsquigarrow$ the bounds are several orders of magnitude above the error...



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One can change the metric to get

$$\left\| \boldsymbol{M}^{1/2} \Pi_P (P - P_*) \right\|_{\mathrm{F}}$$
$$\leqslant \left\| \boldsymbol{M}^{1/2} (\boldsymbol{\Omega}_* + \boldsymbol{K}_*)^{-1} \boldsymbol{M}^{1/2} \right\|_{\mathrm{op}} \left\| \boldsymbol{M}^{-1/2} R(P) \right\|_{\mathrm{F}}.$$

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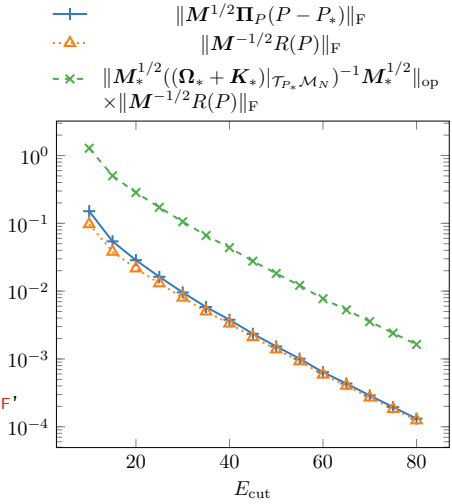
$\rightsquigarrow$ asymptotically $\left\| \boldsymbol{M}^{-1/2} R(P) \right\|_{\mathrm{F}} \sim \left\| \boldsymbol{M}^{1/2} \Pi_P (P - P_*) \right\|_{\mathrm{F}}$, though not upper bound nor guaranteed. The same holds for $\left\| \boldsymbol{M}^{-1} R(P) \right\|_{\mathrm{F}} \sim \| P - P_* \|_{\mathrm{F}}$.



Error bounds for the forces

Forces are decomposed into two components (local and non-local)[3].

Local forces: Let $F^{\mathrm{loc}}_{j,\alpha}(P)$ be the local forces on atom j in direction α. It holds (at first order):

$$F^{\mathrm{loc}}_{j,\alpha}(P) - F^{\mathrm{loc}}_{j,\alpha}(P_*) = \mathrm{d}F^{\mathrm{loc}}_{j,\alpha}(P) \cdot \Pi_P(P - P_*);$$

$$\left|F^{\mathrm{loc}}_{j,\alpha}(P) - F^{\mathrm{loc}}_{j,\alpha}(P_*)\right| \leqslant \left\|\mathrm{d}F^{\mathrm{loc}}_{j,\alpha}(P)\right\|_{\mathcal{T}_P\mathcal{M}_N \to \mathbb{R}} \|P - P_*\|_{\mathrm{F}}.$$

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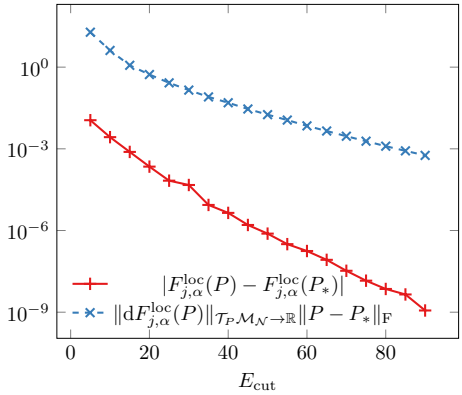

$\rightsquigarrow$ several orders of magnitude above !

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Total forces : Combining local and nonlocal forces on all atoms, we have $F(P) \in \mathbb{R}^{3N_{\#\mathrm{atoms}}}$ and

$$F(P) - F(P_*) = \mathrm{d}F(P) \cdot \Pi_P(P - P_*).$$

$\rightsquigarrow$ What happens if we directly replace $\Pi_P(P - P_*)$ by $\boldsymbol{M}^{-1}R(P)$ in $\mathrm{d}F(P) \cdot \Pi_P(P - P_*)$?

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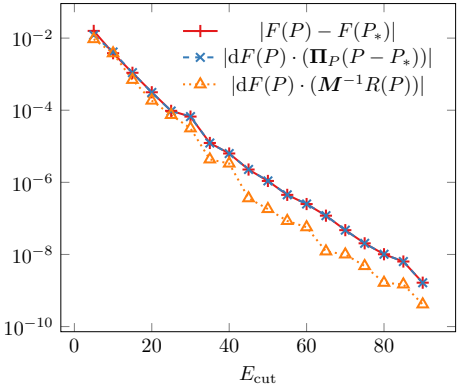

$\rightsquigarrow$ linearization quickly valid;
$\rightsquigarrow$ even if $\Pi_P(P - P_*)$ and $\boldsymbol{M}^{-1}R(P)$ are asymptotically equivalent, orange and blue do not match.

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Frequency splitting

Let $P \in \mathcal{M}_N$, then $\mathcal{T}_P\mathcal{M}_N$ can be split into low and high frequencies. More precisely, given $E_{\mathrm{cut}} < E_{\mathrm{cut,ref}}$, we have

$$\begin{array}{ccccc} \mathcal{T}_P\mathcal{M}_N & = & \Pi_{E_{\mathrm{cut}}}\mathcal{T}_P\mathcal{M}_N & \oplus & \Pi^{\perp}_{E_{\mathrm{cut}}}\mathcal{T}_P\mathcal{M}_N \\ \cup & & \cup & & \cup \\ X & = & X_1 & + & X_2 \\ \updownarrow & & \updownarrow & & \updownarrow \\ \psi & = & \psi_1 & + & \psi_2 \end{array}$$

with $\psi_1 \in \mathcal{X}_{E_{\mathrm{cut}}}$, $\psi_2 \in \mathcal{X}^{\perp}_{E_{\mathrm{cut}}}$ and $\mathcal{X}_{E_{\mathrm{cut,ref}}} = \mathcal{X}_{E_{\mathrm{cut}}} \oplus \mathcal{X}^{\perp}_{E_{\mathrm{cut}}}$.

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If P is a solution of the variational problem for a given E_{cut}, then $R(P), \boldsymbol{M}^{-1}R(P) \in \Pi^{\perp}_{E_{\mathrm{cut}}}\mathcal{T}_P\mathcal{M}_N$ (not exactly true in practice because of numerical quadrature errors due to exchange-correlation terms.).

Let us analyze in details the computation of $F^{\mathrm{loc}}_{j,\alpha}(P)$: $F^{\mathrm{loc}}_{j,\alpha}(P) = -\operatorname{Tr}\left(\dfrac{\partial V_{\mathrm{loc}}}{\partial R_{j,\alpha}} P\right)$ so that computing $\mathrm{d}F^{\mathrm{loc}}_{j,\alpha}(P) \cdot X$ for $X \in \mathcal{T}_P\mathcal{M}_N$ reduces to the scalar product of X against $\Pi_P \dfrac{\partial V_{\mathrm{loc}}}{\partial R_{j,\alpha}}$.

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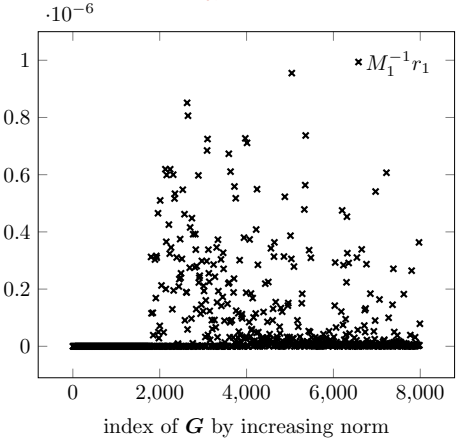
- $\boldsymbol{M}^{-1}R(P)$ is high frequencies;
- $\Pi_P(P - P_*)$ is mainly high frequencies but with low frequencies components;
- $\Pi_P \dfrac{\partial V_{\mathrm{loc}}}{\partial R_{j,\alpha}}$ is mainly low frequencies.



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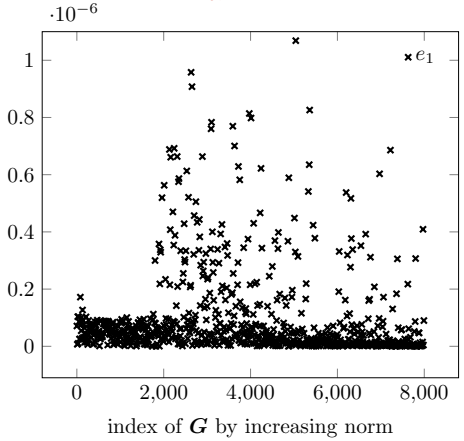

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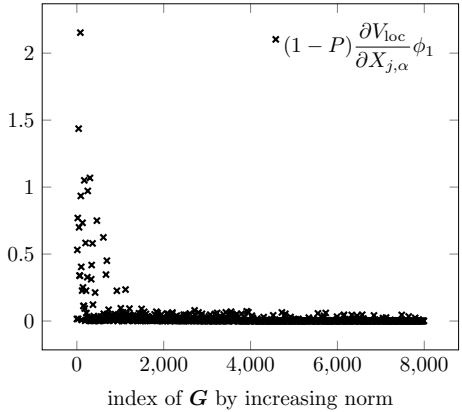

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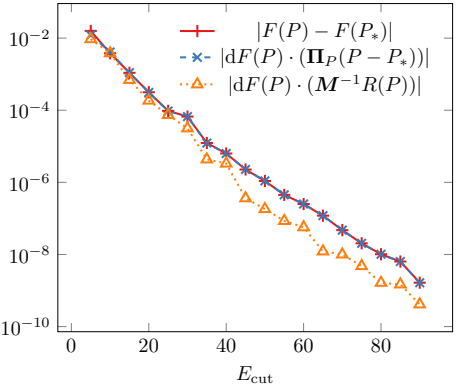
$\rightsquigarrow$ orange and blue do not match because the error and the residual don't have the same support in frequencies, even if $\left\|\boldsymbol{M}^{-1}R(P)\right\|_{\mathsf{F}} \sim \|\Pi_P(P - P_*)\|_{\mathsf{F}}$ asymptotically.



Enhanced error bounds

We decompose the error/residual relation onto $\Pi_{E_{\mathrm{cut}}}\mathcal{T}_P\mathcal{M}_N \oplus \Pi_{E_{\mathrm{cut}}}\mathcal{T}_P\mathcal{M}_N^{\perp}$ to get

$$\begin{bmatrix} (\boldsymbol{\Omega}+\boldsymbol{K})_{11} & (\boldsymbol{\Omega}+\boldsymbol{K})_{12} \\ (\boldsymbol{\Omega}+\boldsymbol{K})_{21} & (\boldsymbol{\Omega}+\boldsymbol{K})_{22} \end{bmatrix} \begin{bmatrix} P_1 - P_{*1} \\ P_2 - P_{*2} \end{bmatrix} = \begin{bmatrix} R_1 \\ R_2 \end{bmatrix}.$$

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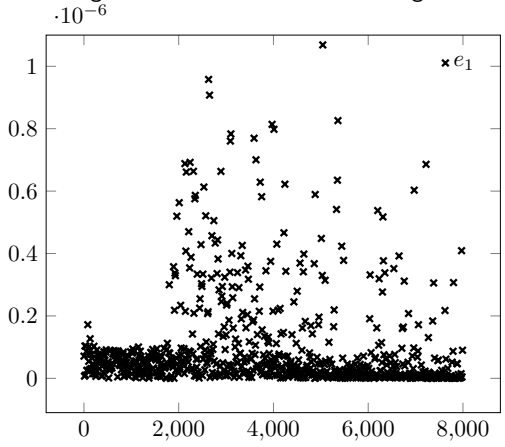
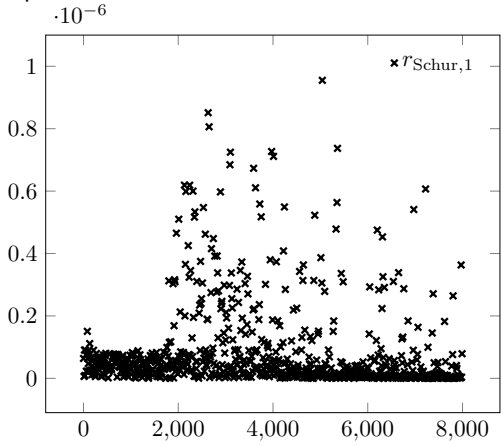
This yields a new residual, which requires only an inversion on the coarse grid $\mathcal{X}_{E_{\mathrm{cut}}}$ ($\boldsymbol{M}_{22}$ being easy to invert):

$$\boxed{R_{\mathrm{Schur}}(P) = \begin{bmatrix} (\boldsymbol{\Omega}+\boldsymbol{K})_{11}^{-1}\left(R_1 - (\boldsymbol{\Omega}+\boldsymbol{K})_{12}\,\boldsymbol{M}_{22}^{-1}R_2\right) \\ \boldsymbol{M}_{22}^{-1}R_2 \end{bmatrix}}.$$

We managed to recover what was missing in the low frequencies.





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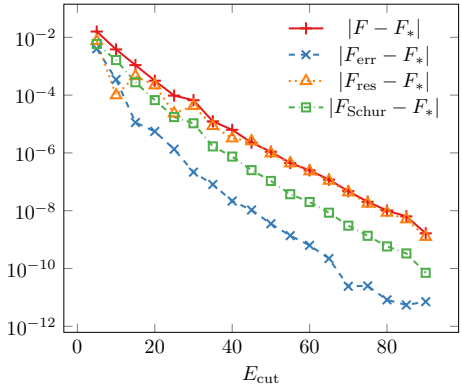

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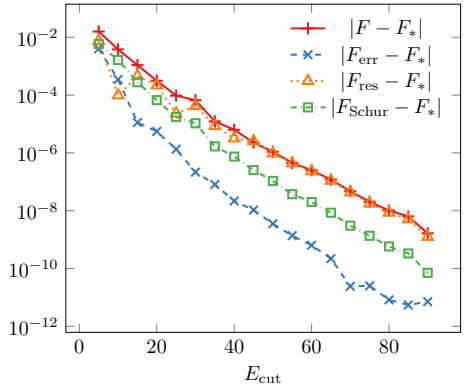
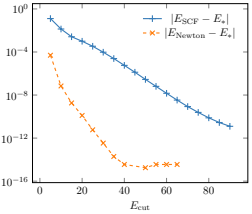
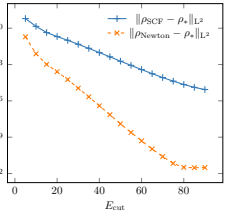
$\rightsquigarrow$ we win about one order of magnitude in the approximation of the error of the forces $F - F_*$.

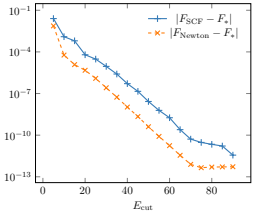


Numerical examples

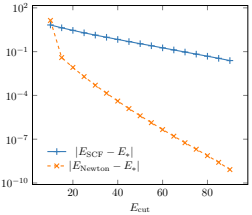
GaAs

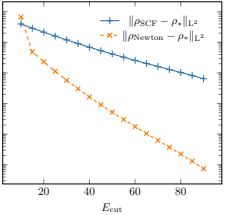


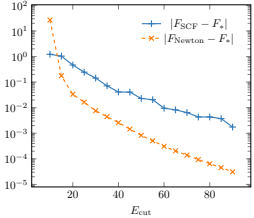




TiO_2







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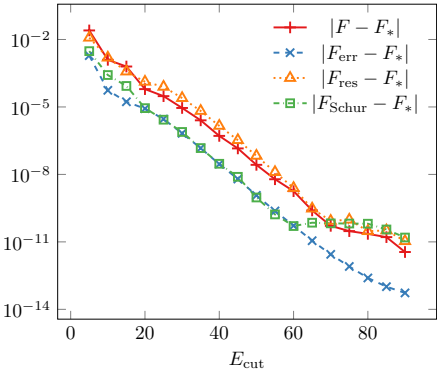

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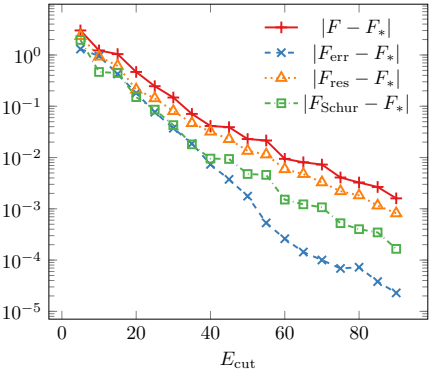

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Preprint with more details
`https://hal.inria.fr/hal-03408321`

Merci!

Let's finish with a glimpse of Toulouse!