Limits and Bounds – as Good as it Sounds

Regularised Density-Potential Inversion for Periodic Systems

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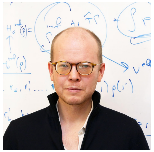
Acknowledgements

In collaboration with Oliver M. Bohle[1,2], Maryam Lotfigolian[1], André Laestadius[1,2] & Erik I. Tellgren[2].

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Motivation

- Success of DFT depends on exchange-correlation approximations

Interacting electrons

$$H(v) = -\frac{1}{2}\sum_i \nabla_i^2 + \sum_i v_{\text{ext}}(\mathbf{r}_i) + \sum_{i,j} w(\mathbf{r}_i - \mathbf{r}_j)$$

ρ_{gs}

Non-interacting electrons (KS system)

$$H_{\text{KS}} = -\frac{1}{2}\sum_i \nabla_i^2 + \sum_i \underbrace{[v_{\text{ext}}(\mathbf{r}_i) + v_{\text{H}}(\mathbf{r}_i) + v_{\text{xc}}(\mathbf{r}_i)]}_{v_{\text{s}}(\mathbf{r}_i)}$$

- Inverse Kohn–Sham: given ground-state density, find effective potential
- Moreau–Yosida regularisation

$$F(\rho) = \sup_{v\in X^*}\{E(v) - \langle v,\rho\rangle\} \quad \longrightarrow \quad F^{\varepsilon}(\rho) = \sup_{v\in X^*}\left\{E(v) - \frac{\varepsilon}{2}\|v\|_{X^*}^2 - \langle v,\rho\rangle\right\}$$

- Periodic systems

Theory

Periodic DFT Framework

- Periodic function spaces over the unit cell $\mathcal{C}$

$$\rho \in X = H^{-1}(\mathcal{C}), \quad v \in X^* = H^1(\mathcal{C}) \tag{1}$$

- Born–von Kármán boundary conditions
- Hamiltonian

$$H_\lambda(v) = -\frac{1}{2}\sum_{i=1}^{n}\nabla_i^2 + \sum_{i=1}^{n} v_{\text{ext}}(\mathbf{r}_i) + \lambda \sum_{i=1}^{n}\sum_{j=1}^{M} w(\mathbf{r}_i - \mathbf{r}_j) = T + V + \lambda W \tag{2}$$

- Yukawa interaction w
- Explicit form of the duality map $J : X \to X^*$

$$J(\widehat{\rho})(\mathbf{G}) = \frac{\widehat{\rho}(\mathbf{G})}{\gamma^2 + G^2}, \quad \mathbf{G} \in \mathcal{RL} \tag{3}$$

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Moreau–Yosida Regularisation

- Constrained-search universal functional

$$F_\lambda(\rho) = \inf_{\Gamma\mapsto\rho} \mathrm{Tr}\,\Gamma H_\lambda(0)$$
$$E_\lambda(v) = \inf_{\rho\in X} \left(F_\lambda(\rho) + \langle v, \rho\rangle\right) \tag{4}$$

- Moreau–Yosida (MY) regularisation

$$F_\lambda^\varepsilon(\rho) = \inf_{\sigma\in X} \left(F_\lambda(\sigma) + \tfrac{1}{2\varepsilon}\|\sigma-\rho\|_X^2\right), \quad \varepsilon > 0 \tag{5}$$

- Proximal density

$$\rho^\varepsilon = \operatorname{prox}_{\varepsilon F}(\rho) = \operatorname*{argmin}_{\sigma\in X} \left(F_\lambda(\sigma) + \tfrac{1}{2\varepsilon}\|\sigma-\rho\|_X^2\right) \tag{6}$$

MY inverse Kohn–Sham

- Model functional

$$F^{\mathrm{mod}}_{\alpha,\xi}(\rho) = T_{\mathrm{s}}(\rho) + \alpha\langle v_{\mathrm{ext}}, \rho\rangle + \xi E_{\mathrm{H}}(\rho,\rho), \quad T_{\mathrm{s}}(\rho) = F_{\lambda=0}(\rho) \tag{7}$$

- Effective KS potential

$$v^{\varepsilon}_{\mathrm{s}} = \alpha v_{\mathrm{ext}} + \xi v_{\mathrm{H}}(\rho^{\varepsilon}) + \frac{1}{\varepsilon} J(\rho^{\varepsilon} - \rho_{\mathrm{ref}}), \quad \rho^{\varepsilon} = \operatorname*{prox}_{\varepsilon F^{\mathrm{mod}}_{\alpha,\xi}}(\rho_{\mathrm{ref}}) \tag{8}$$

- *The limit*

$$\lim_{\varepsilon\to 0^+} \rho^{\varepsilon} = \rho_{\mathrm{ref}}, \quad \lim_{\varepsilon\to 0^+} v^{\varepsilon}_{\mathrm{s}} = v_{\mathrm{s}}, \tag{9}$$

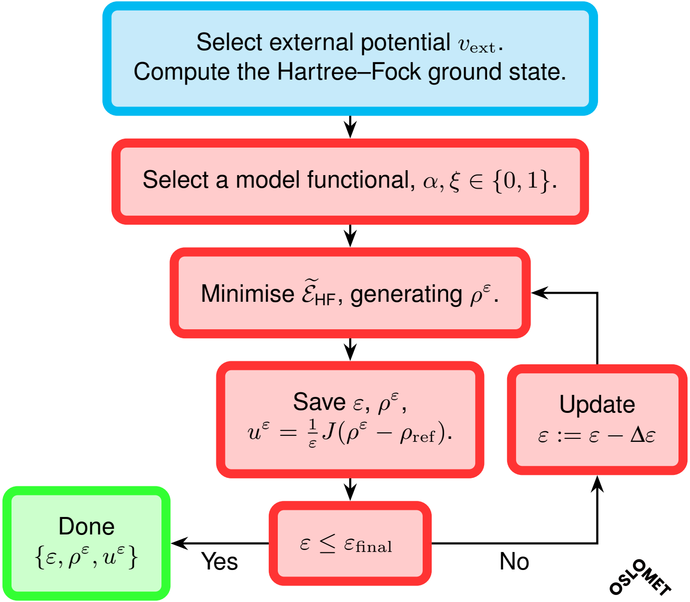
Inversion Scheme

- Main ingredients:
  - Compute the proximal point
  - Generate results for a suitable sequence of ε
- Modified Hartee–Fock energy

$$\widetilde{\mathcal{E}}_{\mathrm{HF}}(D) = \mathrm{Tr}((T + \alpha v)D) + \lambda_{\mathrm{H}}\mathcal{E}_{\mathrm{H}}(D) - \lambda_{\mathrm{x}}\mathcal{E}_{\mathrm{x}}(D) + \frac{1}{2\varepsilon}\|\rho_D - \rho_{\mathrm{ref}}\|_X^2 \tag{10}$$

  - Forward: $\alpha = 1$, $v = v_{\mathrm{ext}}$, $\lambda_{\mathrm{H}} = \lambda_{\mathrm{x}} = 1$, and $1/\varepsilon = 0 \quad \longrightarrow \quad \rho_{\mathrm{ref}}$
  - Inverse: $\lambda_{\mathrm{x}} = 0$, $\lambda_{\mathrm{H}} = \xi$, and $v = v_{\mathrm{ext}} \quad \longrightarrow \quad v_{\mathrm{s}}$

Inversion Scheme



Results

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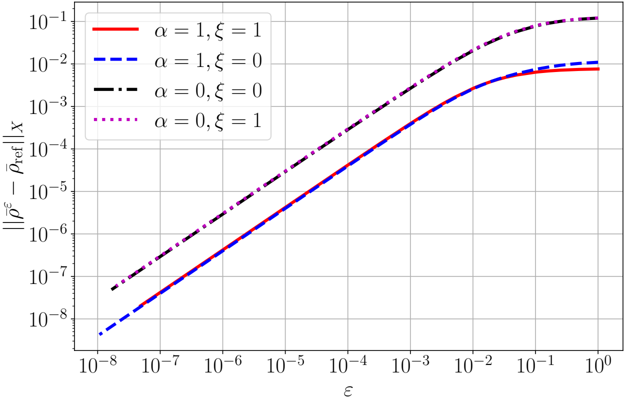
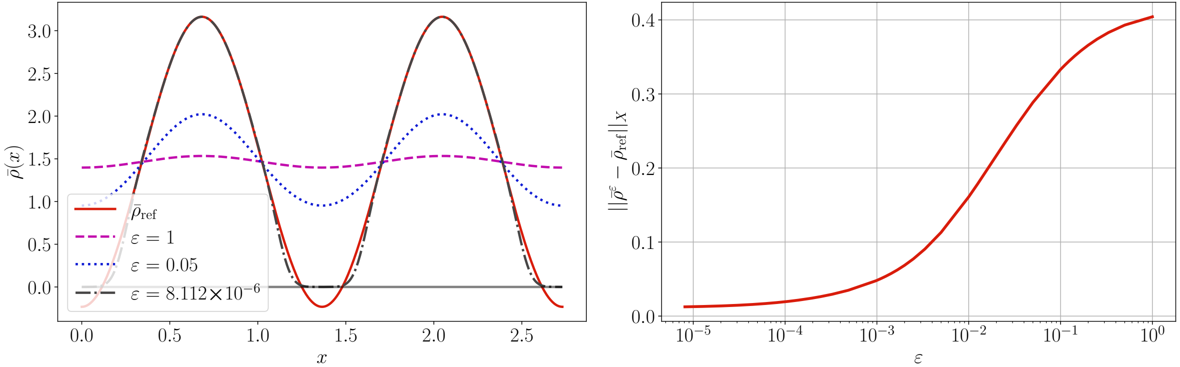
Results

- 1D periodic Hartree–Fock model
- Exact exchange inversion
- Proof-of-principle for extended systems



Non-N-representable Density



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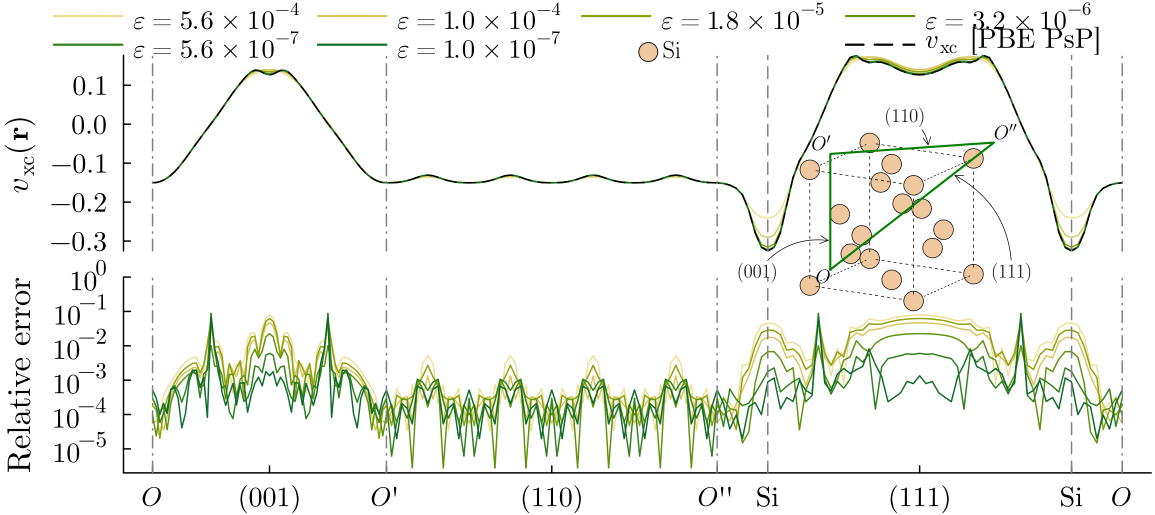
Bulk Silicon



Implementation: github.com/mfherbst/supporting-my-inversion

Error Analysis

Error Bounds

- Consider inexact references

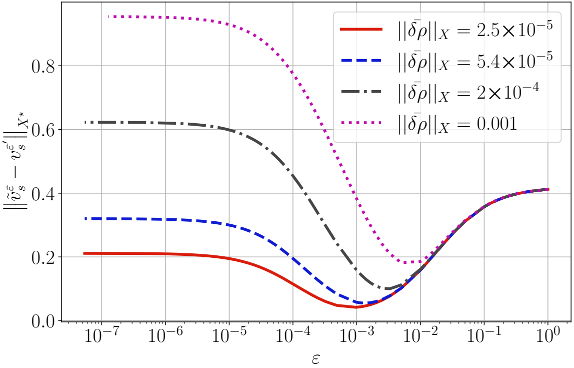
$$\widetilde{\rho}_{\mathrm{ref}} = \rho_{\mathrm{ref}} + \Delta\rho \tag{11}$$

- Gives $\widetilde{v}^{\varepsilon}_{\mathrm{xc}}$ instead of the true $v^{\varepsilon}_{\mathrm{xc}}$
- Bound on the total error

$$\begin{aligned} &\|v_{\mathrm{xc}} - \widetilde{v}^{\varepsilon}_{\mathrm{xc}}\|_{X^*} \\ &\quad\leq \|v_{\mathrm{xc}} - v^{\varepsilon}_{\mathrm{xc}}\|_{X^*} + \|v^{\varepsilon}_{\mathrm{xc}} - \widetilde{v}^{\varepsilon}_{\mathrm{xc}}\|_{X^*} \\ &\quad\leq \|v_{\mathrm{xc}} - v^{\varepsilon}_{\mathrm{xc}}\|_{X^*} + \frac{C}{\varepsilon}\|\Delta\rho\|_X \end{aligned} \tag{12}$$

- Optimal value of ε?

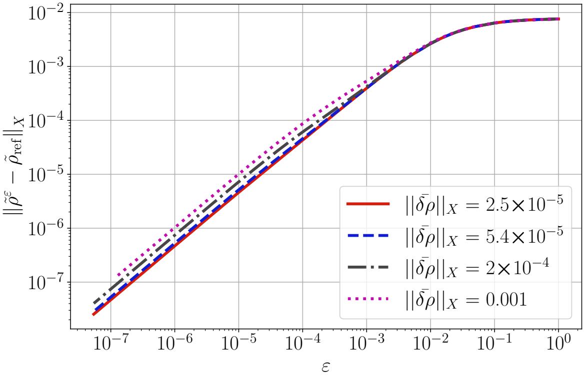
Convergence of potential with Fourier mode cut-off



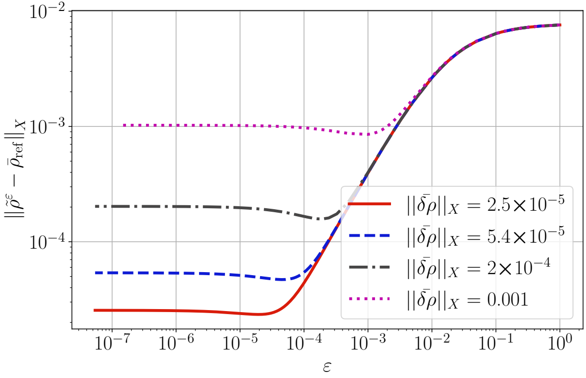
Fourier Mode Cut-Off

$$\widetilde{\rho}_{\text{ref}}(\mathbf{G}) = \rho_{\text{ref}}(\mathbf{G}) + \delta\rho(\mathbf{G}) = \begin{cases} \rho_{\text{ref}}(\mathbf{G}), & \text{if } |\mathbf{G}| \leq G_{\text{trunc}}, \\ 0, & \text{otherwise.} \end{cases} \tag{13}$$

Distance to perturbed density

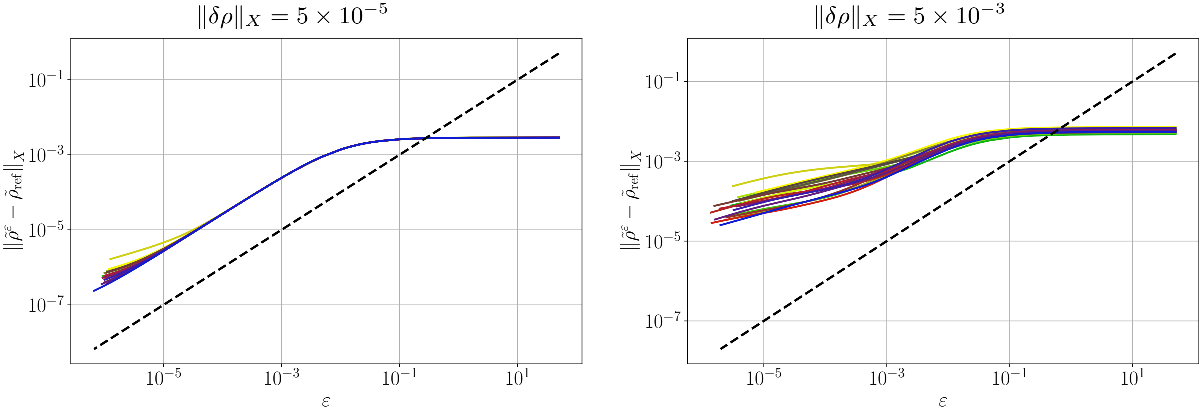


Distance to unperturbed density



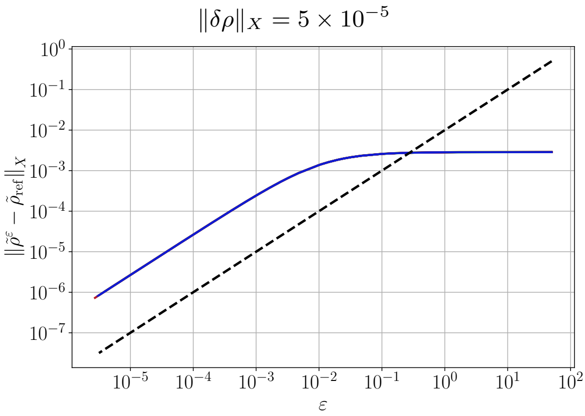
Normally Distributed Noise

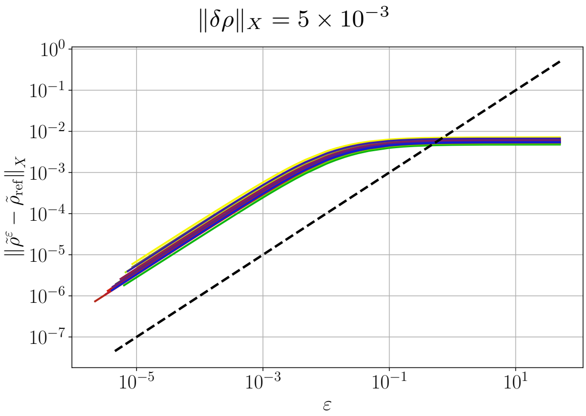
- Perturbations $\delta\rho(\mathbf{G})$ drawn from $\mathcal{N}(0, \sigma_G)$
- Variance $\sigma_G^2 \propto G^{-2}$



Normally Distributed Noise

- More carefully selected noise model





Outlook

Outlook

- Beyond exact exchange
- Higher-dimensional periodic systems
- Theoretical work and development of software
- More physical systems

⋮

- Improved exchange-correlation functional development (?)

References

[1] Bohle, O. M. *et al.*, *Regularized density-potential inversion for periodic systems: Application to exact exchange in one dimension*, *J. Chem. Phys.* **164** (2026) 064104.

[2] Herbst, M. F. *et al.*, *Kohn-Sham inversion with mathematical guarantees*, *Phys. Rev. B* **111** (2025) 205143.

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[4] Herbst, M. F., *mfherbst/supporting-my-inversion: v0.1.0*, 2025.

Thank you!

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Perturbations for Bulk Silicon