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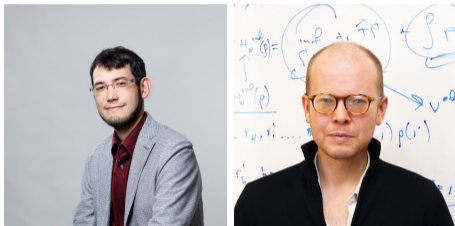
Kohn–Sham Inversion with Mathematical Guarantees

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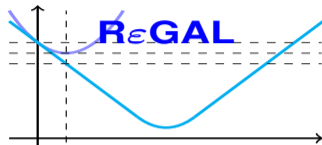
Acknowledgements

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Outline

1 The Kohn–Sham Problem

2 Moreau–Yosida Regularisation

3 The Inversion Scheme

- The Exchange-Correlation Potential
- The Inversion Algorithm

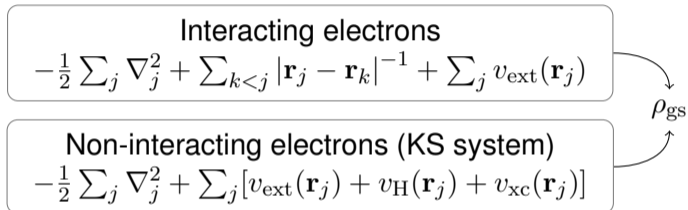
4 Results

- Example: Bulk Silicone
- Error Bounds

5 Conclusions

6 References

The Kohn–Sham Approach



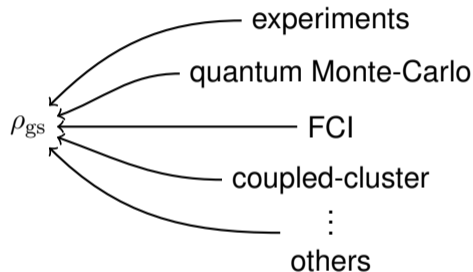
the exchange correlation potential, $v_{\text{xc}} \leftarrow$ unknown

The forward problem: given a v_{xc} , what is the ρ_{gs} ?

The Inverse Kohn–Sham Problem

The reverse problem:

Given a ρ_{gs} , what is the v_{xc} ?



Approach to understanding the *universal density functional* \mathcal{F}

Road to new approximations of \mathcal{F}

Moreau–Yosida Regularisation

Moreau–Yosida Regularisation

Densities: $\rho \in \mathcal{D}$

Potentials: $v \in \mathcal{V} = \mathcal{D}^*$

Definition

Let \mathcal{D} be uniformly convex and $\mathcal{F} : \mathcal{D} \rightarrow \mathbb{R}$ convex and lower semicontinuous functional. For some $\varepsilon > 0$, the *Moreau–Yosida regularisation* of \mathcal{F} is

$$\mathcal{F}^\varepsilon(\rho) = \inf_{\sigma \in \mathcal{D}} \left\{ \mathcal{F}(\sigma) + \frac{1}{2\varepsilon} \|\sigma - \rho\|_{\mathcal{D}}^2 \right\}.$$

An infimal convolution

A Lossless Regularisation

The regularised energy

$$E^\varepsilon(v) = \inf_{\rho \in \mathcal{D}} \{ \mathcal{F}^\varepsilon(\rho) + \langle v, \rho \rangle \}$$

Exact ground-state energy

$$E(v) = E^\varepsilon(v) + \frac{\varepsilon}{2} \|v\|_{\mathcal{V}}^2.$$

The regularisation retains the ground state energy

Consequence of infimal convolution and $E(\text{concave}) \leftrightarrow \mathcal{F}(\text{convex})$.

The Inversion Scheme

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Periodic Systems

$$\mathcal{D} = H_{\text{per}}^{-1}(\Omega, \mathbb{C}) \quad \mathcal{V} = \mathcal{D}^* = H_{\text{per}}^1(\Omega, \mathbb{C})$$

Periodic Sobolev spaces [1]

$$\|u\|_{H_{\text{per}}^s}^2 = \sum_{\mathbf{G}} (1 + |\mathbf{G}|^2)^s |\hat{u}_{\mathbf{G}}|^2$$

The *duality mapping* $J : \mathcal{D} \rightarrow \mathcal{V}$,

$$J[\rho](\mathbf{r}) = (\Phi * \rho)(\mathbf{r}) = \int_{\mathbb{R}^3} \frac{\rho(\mathbf{x})}{4\pi|\mathbf{r} - \mathbf{x}|} e^{-|\mathbf{r} - \mathbf{x}|} d\mathbf{x}$$

Fix $\rho_{\text{gs}} \in \mathcal{D}$ and

$$\mathcal{F}(\rho) = T(\rho) + E_{\text{H}}(\rho) + \int_{\Omega} v_{\text{ext}} \rho$$

The Exchange-Correlation Potential

Minimise

$$\mathcal{E}(\rho; \rho_{\text{gs}}) = \mathcal{F}(\rho) + \frac{1}{2\varepsilon} \|\rho - \rho_{\text{gs}}\|_{\mathcal{D}}^2 \quad \text{over } \rho \in \mathcal{D} \quad (1)$$

Proximal density

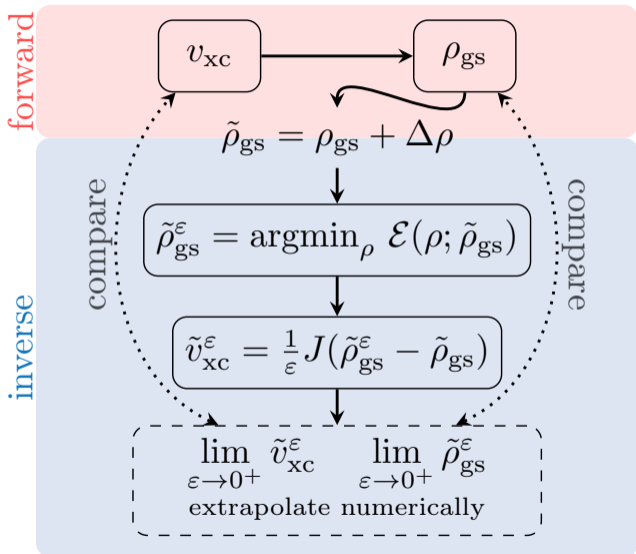
$$\rho_{\text{gs}}^\varepsilon = \underset{\rho}{\operatorname{argmin}} \mathcal{E}(\rho, \rho_{\text{gs}})$$

is the unique minimiser of $\mathcal{E}(\rho; \rho_{\text{gs}})$ [2]

The exchange-correlation potential [3, 4]

$$\begin{aligned} v_{\text{xc}}(\mathbf{r}) &= \lim_{\varepsilon \rightarrow 0^+} \frac{1}{\varepsilon} J[\rho_{\text{gs}}^\varepsilon - \rho_{\text{gs}}](\mathbf{r}) \\ &= \lim_{\varepsilon \rightarrow 0^+} \frac{1}{\varepsilon} \int_{\mathbb{R}^3} \frac{\rho_{\text{gs}}^\varepsilon(\mathbf{x}) - \rho_{\text{gs}}(\mathbf{x})}{4\pi|\mathbf{r} - \mathbf{x}|} e^{-|\mathbf{r} - \mathbf{x}|} d\mathbf{x} \end{aligned}$$

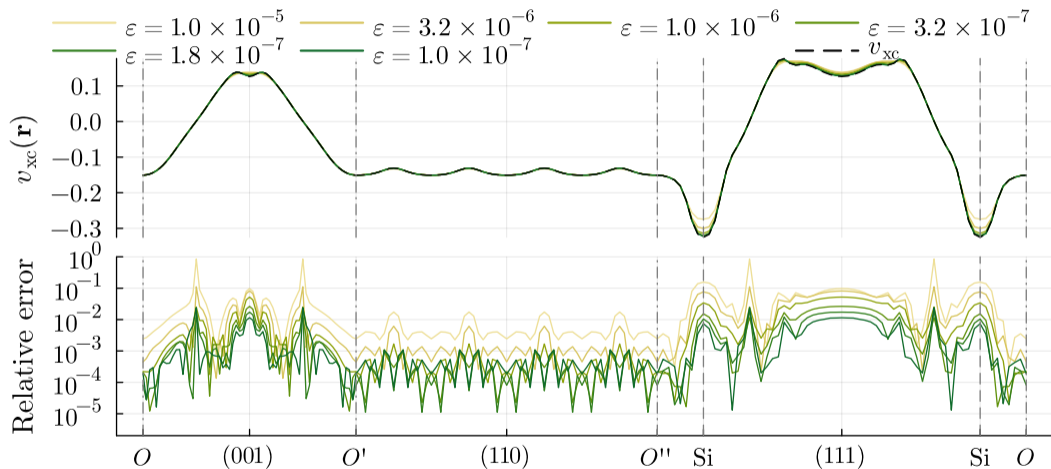
The Inversion Algorithm



Results

- 1 The Kohn–Sham Problem
- 2 Moreau–Yosida Regularisation
- 3 The Inversion Scheme
 - The Exchange-Correlation Potential
 - The Inversion Algorithm
- 4 Results**
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Bulk Silicone



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

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Error Bounds

The proximal mapping $\rho \mapsto \rho^\varepsilon$ is non-expansive,

$$\|\rho^\varepsilon - \tilde{\rho}^\varepsilon\|_{\mathcal{D}} \leq \|\rho - \tilde{\rho}\|_{\mathcal{D}}, \quad \forall \rho, \tilde{\rho} \in \mathcal{D} \quad (2)$$

Consider inexact references:

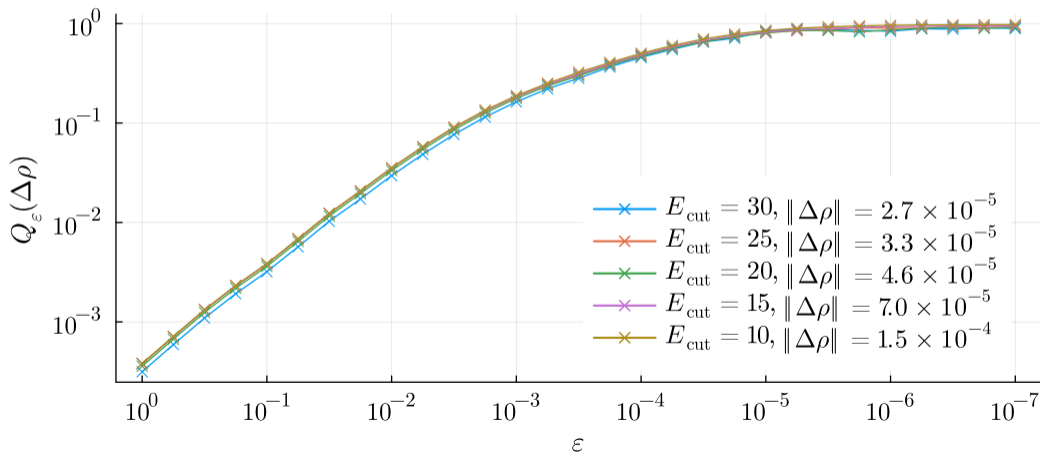
$$\tilde{\rho}_{\text{gs}} = \rho_{\text{gs}} + \Delta\rho$$

The *total error*

$$\|v_{\text{xc}} - \tilde{v}_{\text{xc}}^\varepsilon\|_{\mathcal{V}} \leq \underbrace{\|v_{\text{xc}} - v_{\text{xc}}^\varepsilon\|_{\mathcal{V}}}_{\text{terminating at finite } \varepsilon} + \underbrace{\|v_{\text{xc}}^\varepsilon - \tilde{v}_{\text{xc}}^\varepsilon\|_{\mathcal{V}}}_{\text{using inexact } \rho_{\text{gs}}} \quad (3)$$

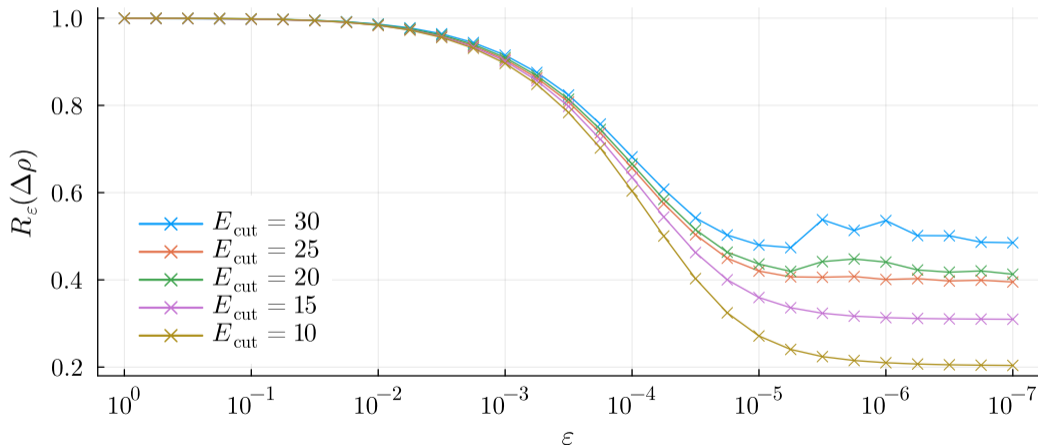
Non-Expansiveness

$$\|\rho_{\text{gs}}^\varepsilon - \tilde{\rho}_{\text{gs}}^\varepsilon\|_{\mathcal{D}} \leq \|\rho_{\text{gs}} - \tilde{\rho}_{\text{gs}}\|_{\mathcal{D}} = \|\Delta\rho\|_{\mathcal{D}} \quad Q_\varepsilon(\Delta\rho) = \frac{\|\rho_{\text{gs}}^\varepsilon - \tilde{\rho}_{\text{gs}}^\varepsilon\|_{\mathcal{D}}}{\|\Delta\rho\|_{\mathcal{D}}}$$



Error Bounds: Inexact Densities

$$\|\tilde{v}_{\text{xc}}^\varepsilon - v_{\text{xc}}^\varepsilon\|_{\mathcal{V}} \leq \frac{1 + Q_\varepsilon(\Delta\rho)}{\varepsilon} \|\Delta\rho\|_{\mathcal{D}} \quad R_\varepsilon(\Delta\rho) = \varepsilon \frac{\|\tilde{v}_{\text{xc}}^\varepsilon - v_{\text{xc}}^\varepsilon\|_{\mathcal{V}}}{\|\Delta\rho\|_{\mathcal{D}}}$$



Conclusions

- Mathematically rigorous inversion scheme
- Rigorous error estimates
- Practical use case of Moreau–Yosida regularisation
- First system studied: bulk silicone
- Implemented using `DFTK.jl` [5]



DFTK

Outlook

- More complicated systems
- Reference densities from other sources
- Further error analysis
- Investigate proximal point algorithms

References I

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Thank you for your attention!

Available on [arXiv:2409.04372](https://arxiv.org/abs/2409.04372)

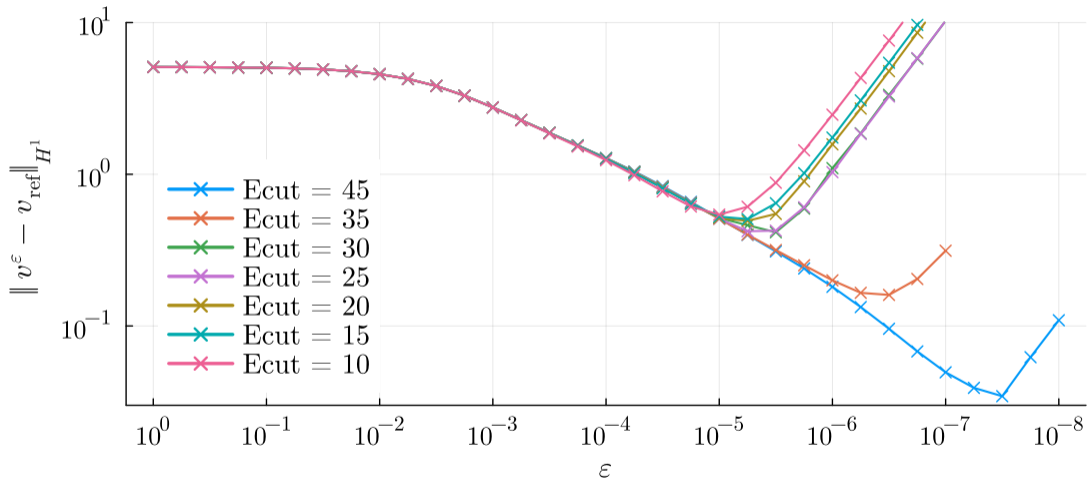
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Motivation: The Differentiability of \mathcal{F}

- The exact universal functional \mathcal{F} is non-differentiable with respect to ρ [6].
- Practical implementations often assume differentiability, e.g., $v_{xc} = \delta E_{xc} / \delta \rho$.
- Regularisation of \mathcal{F} to ensure differentiability.

Convergence: Bulk Silicon



Analytic Error Bounds

Theoretically, the ratios satisfies $\forall \Delta\rho \in \mathcal{D}$

$$0 \leq Q_\varepsilon(\Delta\rho) \leq 1$$

$$0 \leq 1 - Q_\varepsilon(\Delta\rho) \leq R_\varepsilon(\Delta\rho) \leq 1 + Q_\varepsilon(\Delta\rho) \leq 2$$

$$0 \leq S_\varepsilon(\Delta\rho) \leq Q_\varepsilon(\Delta\rho) \leq 1$$

$$\lim_{\varepsilon \rightarrow 0^+} Q_\varepsilon(\Delta\rho) = 1 \quad \text{and} \quad \lim_{\varepsilon \rightarrow 0^+} S_\varepsilon(\Delta\rho) \leq 1$$

Error Bounds: Inexact Densities

$$\|v_{\text{xc}}^\varepsilon - \tilde{v}_{\text{xc}}^\varepsilon - \frac{1}{\varepsilon} J(\Delta\rho)\|_{\mathcal{V}} \leq \frac{1}{\varepsilon} Q_\varepsilon(\Delta\rho) \|\Delta\rho\|_{\mathcal{D}} \quad S_\varepsilon(\Delta\rho) = \varepsilon \frac{\|v_{\text{xc}}^\varepsilon - \tilde{v}_{\text{xc}}^\varepsilon - \frac{1}{\varepsilon} J(\Delta\rho)\|_{\mathcal{V}}}{\|\Delta\rho\|_{\mathcal{D}}}$$

