Smoothing the Landscape:

Moreau–Yosida Regularization in Modern Density-Functional Theory

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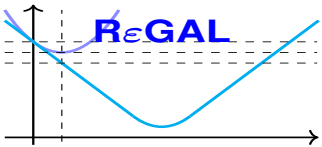

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Outline

1 Introduction to density-functional theory

2 Moreau–Yosida regularisation in DFT

3 Moreau–Yosida inversion scheme

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Introduction to density-functional theory

$$\hat{H}(v) = \overbrace{-\frac{1}{2}\sum_j \nabla_j^2 + \frac{1}{2}\sum_{k\neq j}\frac{1}{|\mathbf{r}_j - \mathbf{r}_k|}}^{\hat{H}_0} + \sum_j v(\mathbf{r}_j) \qquad (\mathbf{r}_1, \ldots, \mathbf{r}_N) \in \mathbb{R}^{3N}$$

$$\psi : \mathbb{R}^{3N} \to \mathbb{C}, \qquad \rho_\psi = N \int_{\mathbb{R}^{3(N-1)}} |\psi|^2 \qquad \langle v,\, \rho\rangle = \int v\rho$$

$$\begin{aligned} E(v) &= \inf_\psi \langle\psi|\hat{H}(v)|\psi\rangle \\ &= \inf_\rho \{F(\rho) + \langle v,\, \rho\rangle\} \end{aligned}$$

$$\begin{aligned} F(\rho) &= \sup_v \{E(v) - \langle v,\, \rho\rangle\} \\ &= \inf_{\Gamma\mapsto\rho} \operatorname{Tr} \hat{H}_0\Gamma \\ &= \inf_{\rho=\sum_j w_j\rho_{\psi_j}} \sum_j w_j \langle\psi_j|\hat{H}_0|\psi_j\rangle \end{aligned}$$

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Hohenberg–Kohn theorem (conjecture?)

Suppose ρ_{gs} is a ground density of $\hat{H}(v_i)$, $i = 1, 2$

$$E(v_i) = \underbrace{\min_{\Gamma \mapsto \rho_{\mathrm{gs}}} \operatorname{Tr} \hat{H}_0 \Gamma}_{\text{independent of } v_i} + \langle v_i, \rho_{\mathrm{gs}} \rangle$$

$\implies$ $\hat{H}(v_1)$ and $\hat{H}(v_2)$ share a ground state ψ $\implies$

$$\text{subtracting} \quad \begin{cases} \hat{H}(v_1)\psi &= E(v_1)\psi \\ \hat{H}(v_2)\psi &= E(v_2)\psi \end{cases} \implies$$

$$(v_1(\mathbf{r}_1) - v_2(\mathbf{r}_1))\psi = \overbrace{\Big(E(v_1) - E(v_2) - \sum_{j=2}^{N} (v_1(\mathbf{r}_j) - v_2(\mathbf{r}_j)) \Big)}^{\text{independent of } x_1} \psi$$

If $\psi \neq 0$ (a.e.), “UCP” in the math. litt., then $v_1 - v_2 =$ constant.

DFT: (F, E) conjugate pair

$$F(\rho) = \sup_{v} \{E(v) - \langle v,\, \rho \rangle\}$$

$$E(v) = \inf_{\rho} \{F(\rho) + \langle v,\, \rho \rangle\}$$

In general: $E(v) - F(\rho) \leq \langle v, \rho \rangle$

and equality for a ground-state pair (v, ρ)

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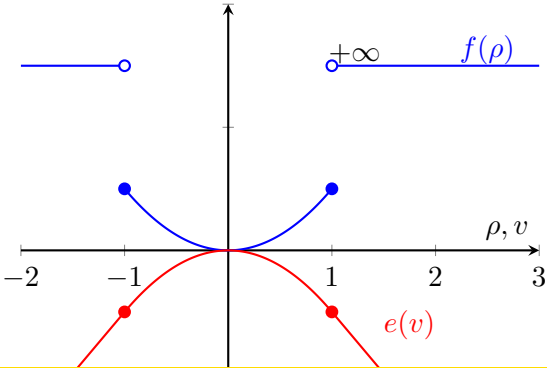
Example: conjugation of a simple function

$$f(\rho) = \begin{cases} \frac{1}{2}\rho^2, & \text{if } |\rho| \le 1, \\ \infty, & \text{if } |\rho| > 1 \end{cases} \quad \Rightarrow \quad e(v) = \inf_{\rho}(f(\rho) + v\rho) = \begin{cases} \frac{1}{2} - v, & \text{if } v > 1, \\ -\frac{1}{2}v^2, & \text{if } |v| \le 1, \\ \frac{1}{2} + v, & \text{if } v < -1. \end{cases}$$



Moreau–Yosida regularisation in DFT

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Moreau–Yosida regularisation

Duality mapping $J_{\text{dual}} : X \to X^*$ (X str. convex and reflex., X^* uni. convex)

Density space: N-rep $\subsetneq X \ni \rho$ Potential space: $X^* \ni v$

Definition

Let $F : X \to \mathbb{R} \cup \{+\infty\}$ convex and lower semicontinous functional. For some $\varepsilon > 0$, the *Moreau–Yosida regularisation* of F is

$$F^\varepsilon(\rho) = \inf_{\sigma \in X} \left\{ F(\sigma) + \tfrac{1}{2\varepsilon} \|\sigma - \rho\|_X^2 \right\}$$

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

Why? $F(\rho)$ in std DFT is **everywhere** discontinuous, P. Lammert [1]
Introduced into DFT by Kvaal et al. [2]

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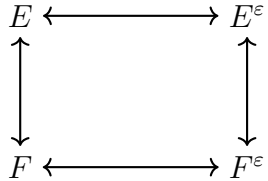
A Lossless Regularisation

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

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



The regularisation retains the ground-state energy and achieves diff.

$$v^\varepsilon(\rho) = -\frac{\delta}{\delta\rho}F^\varepsilon(\rho) = \frac{1}{\varepsilon}J_{\mathrm{dual}}[\rho^\varepsilon - \rho]$$

Quantitative HK:

$$\|v^\varepsilon(\rho_1) - v^\varepsilon(\rho_1)\|_{X^*} \leq \frac{C}{\varepsilon}\|\rho_1 - \rho_2\|_X$$

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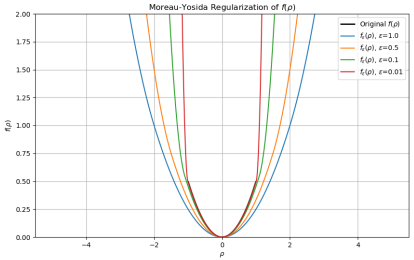
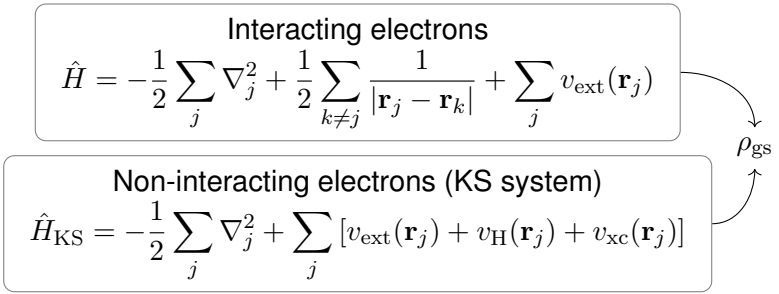
Example: Regularization of a simple function

$$f(\rho) = \begin{cases} \frac{1}{2}\rho^2, & \text{if } |\rho| \le 1, \\ \infty, & \text{if } |\rho| > 1 \end{cases} \quad \Rightarrow \quad f^{\varepsilon}(\rho) = \begin{cases} \frac{1}{2} + \frac{1}{2\varepsilon}(1-\rho)^2, & \text{if } \rho \ge 1+\varepsilon, \\ \frac{\rho^2}{2(1+\varepsilon)}, & \text{if } |\rho| \le 1+\varepsilon, \\ \frac{1}{2} + \frac{1}{2\varepsilon}(1+\rho)^2, & \text{if } \rho \le -1-\varepsilon. \end{cases}$$



The Kohn–Sham Approach



Exchange-correlation potential $v_{\mathrm{xc}} \longleftarrow$ unknown

The forward problem:
Given a v_{ext} and an approximate E_{xc}, i.e., v_{xc}, determine ρ_{gs}

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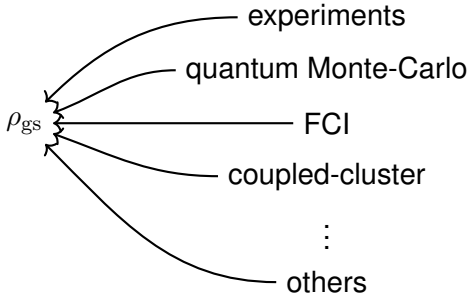
The Inverse Kohn–Sham Problem

The reverse problem:
Given a ρ_{gs}, determine v_{xc}



Why?

Better understanding of $F(\rho)$
Additional constraints for $F(\rho)$ to use when designing approximations

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Moreau–Yosida inversion scheme

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Kohn–Sham model functional

Recall

$$\hat{H}_{\mathrm{KS}} = -\frac{1}{2}\sum_j \nabla_j^2 + \sum_j \overbrace{\left[v_{\mathrm{ext}}(\mathbf{r}_j) + v_{\mathrm{H}}(\mathbf{r}_j) + v_{\mathrm{xc}}(\mathbf{r}_j)\right]}^{v_{\mathrm{KS}}}$$

Given ρ_{gs}, let

$$\mathcal{F}(\rho) = T(\rho) + E_{\mathrm{H}}(\rho) + \int v_{\mathrm{ext}}\rho$$

Minimise

$$\mathcal{E}(\rho\,;\rho_{\mathrm{gs}}) = \underbrace{\mathcal{F}(\rho)}_{\text{no xc}} + \underbrace{\tfrac{1}{2\varepsilon}\|\rho-\rho_{\mathrm{gs}}\|_X^2}_{\text{targets xc as } \varepsilon\to 0+} \quad \text{over} \quad \rho\in X \tag{1}$$

Proximal density, unique minimiser of $\mathcal{E}$:

$$\rho_{\mathrm{gs}}^{\varepsilon} = \operatorname*{argmin}_{\rho\in X} \mathcal{E}(\rho,\rho_{\mathrm{gs}})$$

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The Exchange-Correlation Potential

Since $\mathcal{F}^\varepsilon(\rho)$ is differentiable,

$$v_{\mathrm{xc}}^\varepsilon = -\frac{\delta}{\delta\rho}\mathcal{F}^\varepsilon(\rho_{\mathrm{gs}}) = \frac{1}{\varepsilon}J_{\mathrm{dual}}[\rho_{\mathrm{gs}}^\varepsilon - \rho_{\mathrm{gs}}]$$

Theorem. [3, 4] Exchange-correlation potential obtained from

$$v_{\mathrm{xc}}(\mathbf{r}) = \lim_{\varepsilon\to 0^+} v_{\mathrm{xc}}^\varepsilon = \lim_{\varepsilon\to 0^+} \frac{1}{\varepsilon}J_{\mathrm{dual}}\left[\rho_{\mathrm{gs}}^\varepsilon - \rho_{\mathrm{gs}}\right](\mathbf{r})$$

The form of J_{dual} depends on the choice of X:

Ex1. $X = L^2 \implies J_{\mathrm{dual}} = I$ (van Leeuwen & Baerends)

Ex2. $X =$ homogeneous Sobolev space $\implies J_{\mathrm{dual}}(\rho) = v_{\mathrm{H}}(\rho)$ (ZMP)

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Sketch of proof: By assumption v_{KS} exists s.t. the KS system "gives" ρ_{gs}:

$$\min_{\rho}(T(\rho) + \langle v_{\mathrm{KS}}, \rho\rangle) \iff \underline{\partial} T(\rho_{\mathrm{gs}}) + v_{\mathrm{KS}} \ni 0 \tag{2}$$

$$\min_{\rho}(\mathcal{F}(\rho) + \langle v_{\mathrm{xc}}, \rho\rangle) \iff \underbrace{\boxed{\underline{\partial}\mathcal{F}(\rho_{\mathrm{gs}}) + v_{\mathrm{xc}} \ni 0}}_{(*)} \tag{3}$$

Moreau-Yosida:

$$\min_{\rho}\left\{\mathcal{F}(\rho) + \tfrac{1}{2\varepsilon}\|\rho - \rho_{\mathrm{gs}}\|_{\mathcal{D}}^2\right\} \iff \underbrace{\boxed{\underline{\partial}\mathcal{F}(\rho_{\mathrm{gs}}^{\varepsilon}) + \frac{1}{\varepsilon} J_{\mathrm{dual}}(\rho_{\mathrm{gs}}^{\varepsilon} - \rho_{\mathrm{gs}}) \ni 0}}_{(**)} \tag{4}$$

$(*), (**)$ and $\rho_{\mathrm{gs}}^{\varepsilon} \to \rho_{\mathrm{gs}}$ as $\varepsilon \to 0+ \implies v_{\mathrm{xc}} = \lim_{\varepsilon\to 0+} \frac{1}{\varepsilon} J_{\mathrm{dual}}(\rho_{\mathrm{gs}}^{\varepsilon} - \rho_{\mathrm{gs}})$

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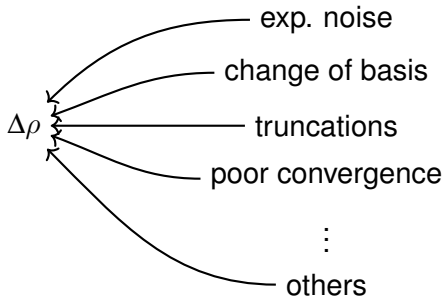
Error Bounds

Inexact density:

$$\widetilde{\rho}_{\mathrm{gs}} = \rho_{\mathrm{gs}} + \Delta\rho$$



The *total error*

$$\|v_{\mathrm{xc}} - \widetilde{v}^{\varepsilon}_{\mathrm{xc}}\|_{X^*} \leq \underbrace{\|v_{\mathrm{xc}} - v^{\varepsilon}_{\mathrm{xc}}\|_{X^*}}_{\text{terminating at finite } \varepsilon} + \underbrace{\|v^{\varepsilon}_{\mathrm{xc}} - \widetilde{v}^{\varepsilon}_{\mathrm{xc}}\|_{X^*}}_{\text{using inexact } \rho_{\mathrm{gs}}} \tag{5}$$

Energy error splitting

Kim, Sim and Burke [5]: Density-driven error

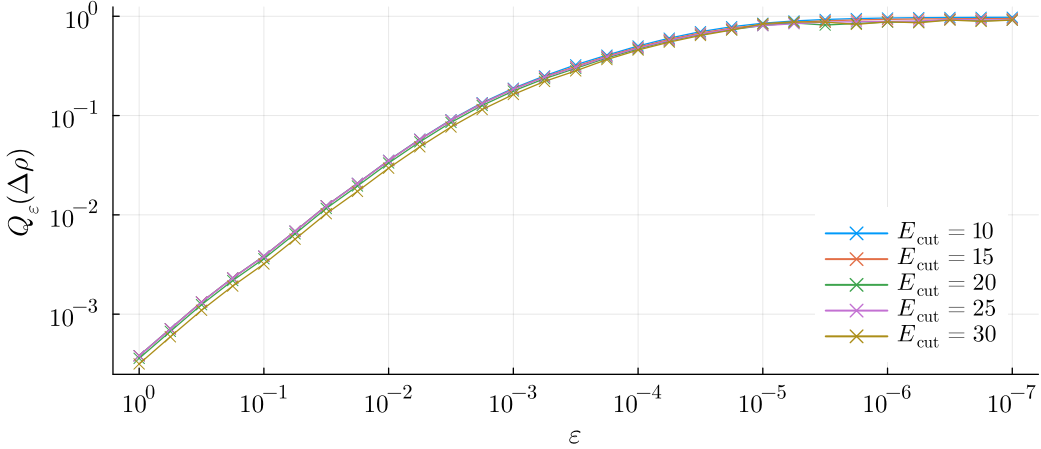
$$\begin{aligned}\Delta E &= E(\rho_{\mathrm{gs}}) - E_{\mathrm{appr}}(\rho_{\mathrm{gs}} + \Delta\rho) \\ &= \underbrace{\big(E(\rho_{\mathrm{gs}}) - E_{\mathrm{appr}}(\rho_{\mathrm{gs}})\big)}_{\text{error from functional}} + \underbrace{\big(E_{\mathrm{appr}}(\rho_{\mathrm{gs}}) - E_{\mathrm{appr}}(\rho_{\mathrm{gs}} + \Delta\rho)\big)}_{\text{error from density}}\end{aligned} \tag{6}$$

We borrow the term “density-driven” when we speak of errors in the potentials caused by inexact densities:

$$\|v_{\mathrm{xc}}^{\varepsilon} - \widetilde{v}_{\mathrm{xc}}^{\varepsilon}\|_{X^*} \leq \frac{1 + Q_{\varepsilon}}{\varepsilon}\|\Delta\rho\|_X$$

Non-Expansiveness of $\rho \longmapsto \rho^{\varepsilon}$

$$\left\|\rho_{\mathrm{gs}}^{\varepsilon} - \widetilde{\rho}_{\mathrm{gs}}^{\varepsilon}\right\|_X \leq \left\|\rho_{\mathrm{gs}} - \widetilde{\rho}_{\mathrm{gs}}\right\|_X = \left\|\Delta\rho\right\|_X \qquad Q_{\varepsilon}(\Delta\rho) = \frac{\left\|\rho_{\mathrm{gs}}^{\varepsilon} - \widetilde{\rho}_{\mathrm{gs}}^{\varepsilon}\right\|_X}{\left\|\Delta\rho\right\|_X}$$



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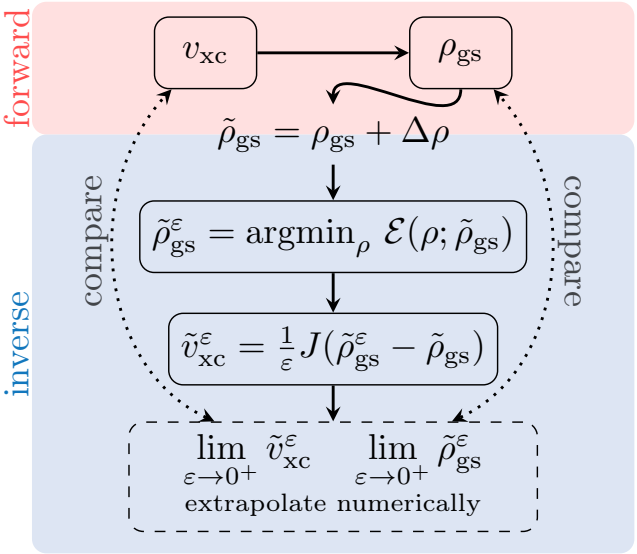
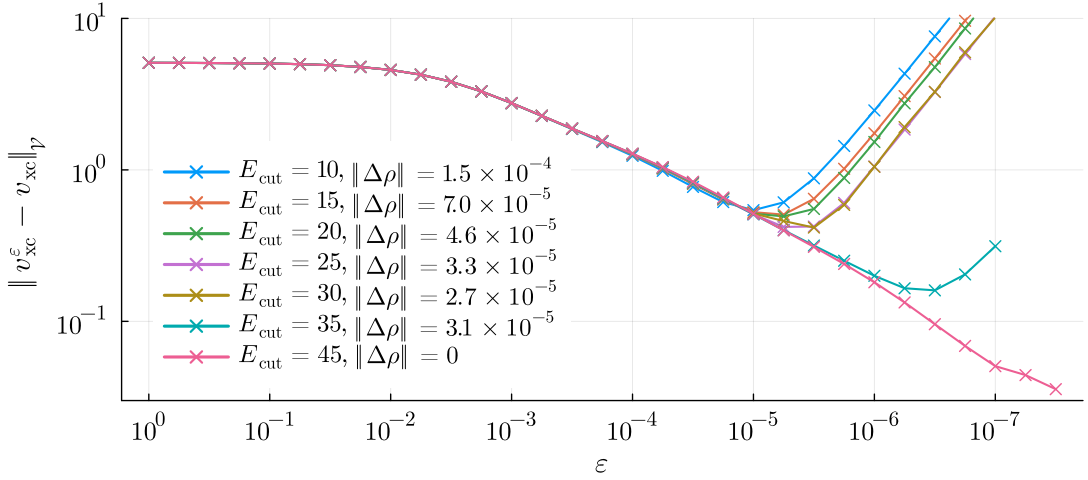
The Inversion Algorithm with error $\Delta\rho$



Convergence: Bulk Silicon



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Conclusion

- Regularization as a bridge between approximate and exact DFT
- Mathematical framework
- Algorithm for determining xc potentials
- Development of approximate functionals

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Thank you for your attention!

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