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Perspectives on Mathematical Pathways in Density-Functional Theory

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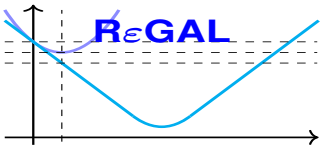

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Outline

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Density-Functional Theory

$$\hat{H}(v) = \overbrace{-\frac{1}{2}\sum_j \nabla_j^2 + \frac{1}{2}\sum_{k\neq j}\frac{1}{|\mathbf{r}_j - \mathbf{r}_k|}}^{\hat{H}_0} + \sum_j v(\mathbf{r}_j) \qquad \left(\hat{T} + \lambda\hat{W} + \hat{V}\right)$$

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

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

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

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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 $\implies$

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

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

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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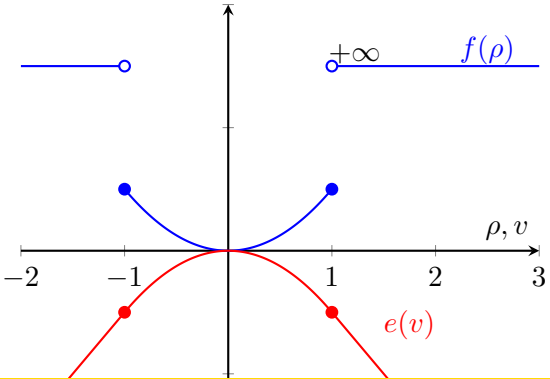
Example: conjugation of a simple function

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



Moreau–Yosida Regularisation

Duality mapping $J_{\mathrm{dual}} : \mathcal{D} \to \mathcal{V}$

Density space: N-rep $\subsetneq \mathcal{D} \ni \rho$ Potential space: $\mathcal{V} = \mathcal{D}^* \ni v$

Definition

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

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

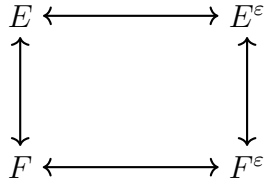
$$\rho^{\varepsilon} = \operatorname*{argmin}_{\sigma \in \mathcal{D}} \left\{ F(\sigma) + \tfrac{1}{2\varepsilon} \|\sigma - \rho\|_{\mathcal{D}}^2 \right\}$$

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

A Lossless Regularisation

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

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



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

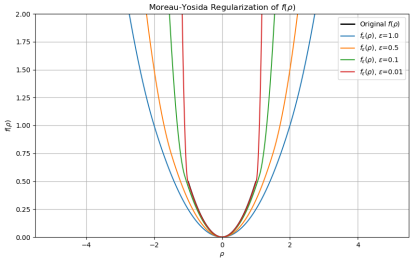
$$v^\varepsilon(\rho) = -\frac{\delta}{\delta\rho}F^\varepsilon(\rho) = \frac{1}{\varepsilon}J_{\mathrm{dual}}[\rho^\varepsilon - \rho]$$

Quantitative HK:

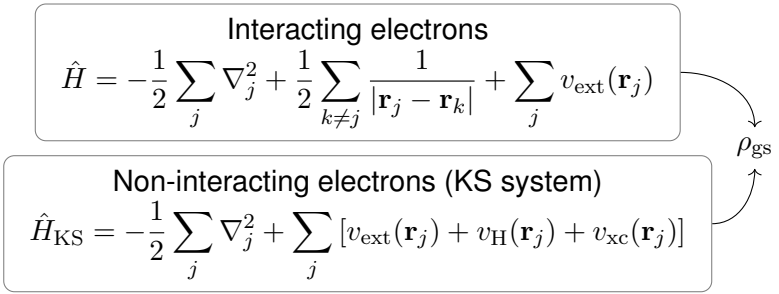
$$\|v^\varepsilon(\rho_1) - v^\varepsilon(\rho_1)\|_{\mathcal{V}} \leq \frac{C}{\varepsilon}\|\rho_1 - \rho_2\|_{\mathcal{D}}$$

Example: Regularization of a simple function

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



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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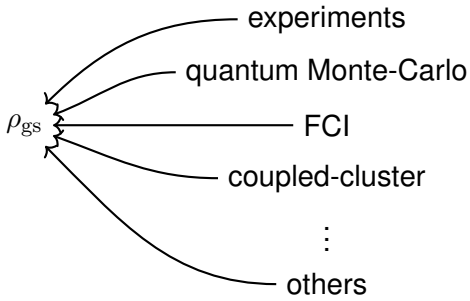
The Inverse Kohn–Sham Problem

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

MY reg. inversion scheme

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“KS” 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

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

Minimise

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

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

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

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 $\mathcal{D}$:
Ex1. $\mathcal{D} = L^2 \implies J_{\mathrm{dual}} = I$ (van Leeuwen & Baerends)
Ex2. $\mathcal{D} =$ 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}:

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

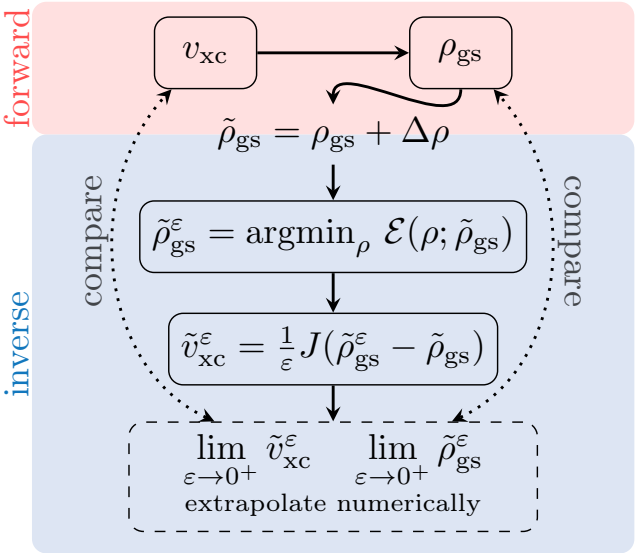
$$\min_{\rho}(\mathcal{F}(\rho) + \langle v_{\mathrm{xc}}, \rho\rangle) \iff \underbrace{\boxed{\underline{\partial}\mathcal{F}(\rho_{\mathrm{gs}}) + v_{\mathrm{xc}} \ni 0}}_{(*)} \tag{3}$$

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

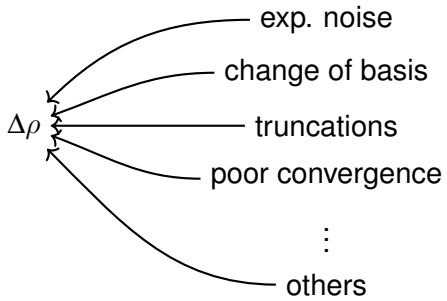
The Inversion Algorithm with error $\Delta\rho$



Error Bounds

Inexact density:

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



The *total error*

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

Energy error splitting

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

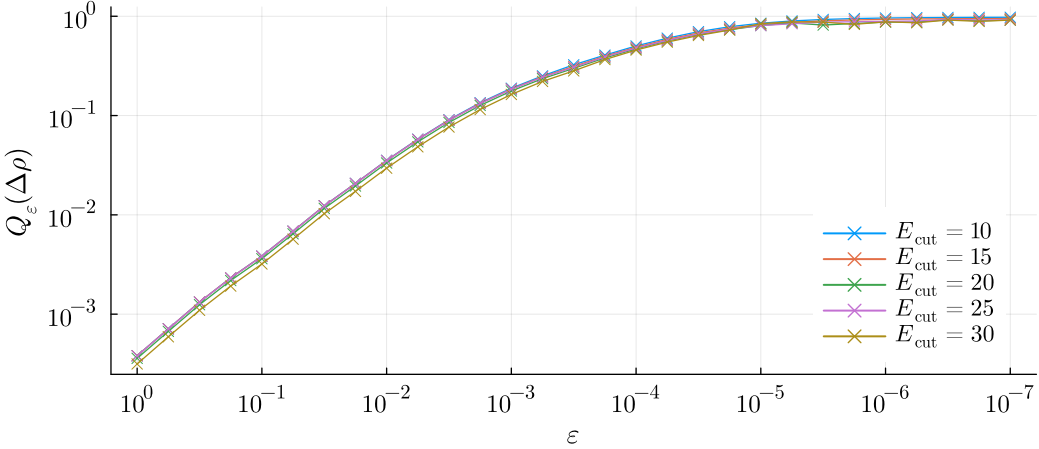
$$\begin{aligned}\Delta E &= E(\rho_{\mathrm{gs}}) - E_{\mathrm{appr}}(\rho_{\mathrm{gs}} + \Delta\rho) \\ &= \underbrace{\big(E(\rho_{\mathrm{gs}}) - E_{\mathrm{appr}}(\rho_{\mathrm{gs}})\big)}_{\text{error from functional}} + \underbrace{\big(E_{\mathrm{appr}}(\rho_{\mathrm{gs}}) - E_{\mathrm{appr}}(\rho_{\mathrm{gs}}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

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

$$\|v_{\mathrm{xc}}^{\varepsilon} - \widetilde{v}_{\mathrm{xc}}^{\varepsilon}\|_{\mathcal{V}} \leq \frac{1 + Q_{\varepsilon}}{\varepsilon} \|\Delta\rho\|_{\mathcal{D}}$$

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

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



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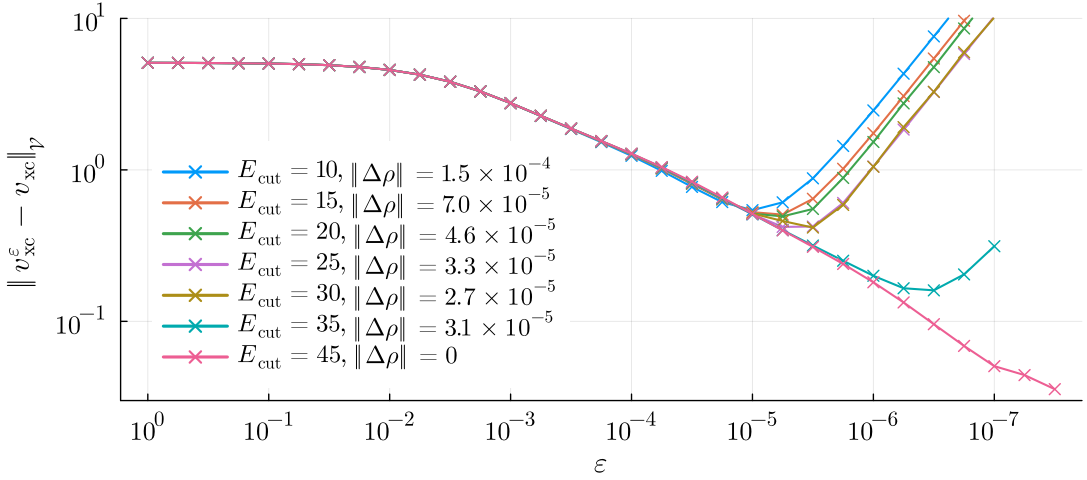
Convergence: Bulk Silicon



MY reg. inversion scheme

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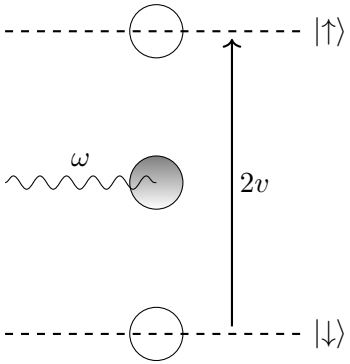
The Dicke Model

- Two physically different subsystems — matter and light
  - N two-level fermionic systems
  - Individually coupled to M modes of a quantized radiation field, described as quantum harmonic oscillators
- Susceptible to a “DFT program”
- We can achieve considerable more mathematically than for standard DFT
  - results concerning v-representability
  - properties of the universal functional



Function spaces

Hilbert space: $\mathcal{H} = \mathcal{H}_{\mathrm{ph}} \otimes \mathcal{H}_{\mathrm{f}}$
$\mathcal{H}_{\mathrm{ph}} = \bigotimes^M L^2(\mathbb{R})$ and $\mathcal{H}_{\mathrm{f}} = \bigotimes^N \mathbb{C}^2 \simeq \mathbb{C}^{2^N}$

$$\mathcal{H} \simeq L^2(\mathbb{R}^M) \otimes \mathbb{C}^{2^N} \simeq L^2(\mathbb{R}^M, \mathbb{C}^{2^N})$$

Inner product $\langle \cdot, \cdot \rangle$ on $L^2(\mathbb{R}^M, \mathbb{C}^{2^N})$,

$$\langle \boldsymbol{\varphi}, \boldsymbol{\psi} \rangle = \sum_{\boldsymbol{\alpha}} \langle \varphi^{\boldsymbol{\alpha}}, \psi^{\boldsymbol{\alpha}} \rangle = \sum_{\alpha_1,\ldots,\alpha_N \in \{+,-\}} \int_{\mathbb{R}^M} \overline{\varphi^{\alpha_1,\ldots,\alpha_N}(\mathbf{x})} \psi^{\alpha_1,\ldots,\alpha_N}(\mathbf{x}) \, \mathrm{d}\mathbf{x},$$

$\psi^{\boldsymbol{\alpha}}$ is the spin projection of ψ corresponding to the eigenvector of the lifted Pauli matrix σ_z^j indexed by the multiindex $\boldsymbol{\alpha} \in \{+,-\}^N$.

Notations

For any $j = 1, \ldots, N$, we have set

$$\sigma_a^j = \mathbb{1} \otimes \ldots \otimes \mathbb{1} \otimes \underbrace{\sigma_a}_{j\text{th}} \otimes \mathbb{1} \otimes \ldots \mathbb{1} \in \mathbb{C}^{2^N \times 2^N},$$

where the Pauli matrices are given by

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma_y = \begin{pmatrix} 0 & -\mathrm{i} \\ \mathrm{i} & 0 \end{pmatrix}, \quad \text{and} \quad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.$$

Vector of lifted Pauli matrices

$$\boldsymbol{\sigma}_a = (\sigma_a^1, \ldots, \sigma_a^N)^\top \in \left(\mathbb{C}^{2^N \times 2^N}\right)^N.$$

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Dicke Hamiltonian

"Internal" part of Hamiltonian $\mathbf{H}_0 : \mathcal{H} \to \mathcal{H}$,

$$\mathbf{H}_0 = (-\Delta + |\mathbf{x}|^2)\mathbb{1}_{\mathbb{C}^{2^N}} + \mathbf{x} \cdot \mathbf{\Lambda}\boldsymbol{\sigma}_z - \mathbf{t} \cdot \boldsymbol{\sigma}_x \tag{7}$$

$\mathbf{\Lambda}\boldsymbol{\sigma}_z$ is to be understood as the M-vector of $2^N \times 2^N$ matrices

$$\mathbf{\Lambda}\boldsymbol{\sigma}_z = \left(\sum_{n=1}^{N} \Lambda_{1n}\sigma_z^n, \ldots, \sum_{n=1}^{N} \Lambda_{Mn}\sigma_z^n \right)^\top.$$

Set $\mathbf{V}(\mathbf{x}) = \left(\mathbf{x} + \frac{1}{2}\mathbf{\Lambda}\boldsymbol{\sigma}_z\right)^2$,

$$\mathbf{H}_0 = -\Delta + \mathbf{V} - \mathbf{t} \cdot \boldsymbol{\sigma}_x - \frac{1}{4}\boldsymbol{\sigma}_z \cdot (\mathbf{\Lambda}^\top \mathbf{\Lambda}\boldsymbol{\sigma}_z),$$

$\mathbf{H}_0$ is bounded from below

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Dicke Hamiltonian

Full Hamiltonian

$$\mathbf{H}(\mathbf{v},\mathbf{j}) = \mathbf{H}_0 + \mathbf{v}\cdot\boldsymbol{\sigma}_z + \mathbf{j}\cdot\mathbf{x} \qquad (8)$$

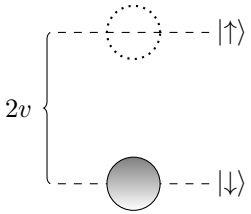
Where the external potentials are

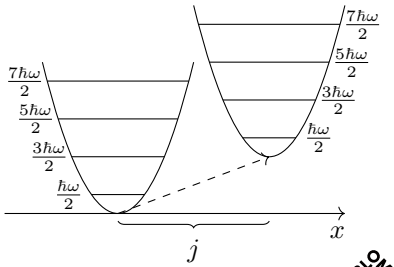
$$\mathbf{v} \in \mathbb{R}^N, \qquad \mathbf{j} \in \mathbb{R}^M$$

Ground-state energy

$$E(\mathbf{v},\mathbf{j}) = \inf_{\substack{\boldsymbol{\psi}\in Q_0 \\ \|\boldsymbol{\psi}\|=1}} \langle \boldsymbol{\psi}, \mathbf{H}(\mathbf{v},\mathbf{j})\boldsymbol{\psi}\rangle \qquad (9)$$

$Q_0 := Q(\mathbf{H}_0) = Q(-\Delta + \mathbf{V})$ form domain of $\mathbf{H}_0$





Internal “density” variables

Definition (Magnetization vector and photon coordinate)

For $\psi \in \mathcal{H}$, we define

$$\boldsymbol{\sigma}_\psi = \langle \psi, \boldsymbol{\sigma}_z \psi \rangle := \begin{pmatrix} \langle \psi, \sigma_z^1 \psi \rangle \\ \vdots \\ \langle \psi, \sigma_z^N \psi \rangle \end{pmatrix} \in [-1,1]^N \subset \mathbb{R}^N$$

$$\boldsymbol{\xi}_\psi = \langle \psi, \mathbf{x}\psi \rangle = \int_{\mathbb{R}^M} \mathbf{x} |\psi(\mathbf{x})|^2 \, \mathrm{d}\mathbf{x} \in \mathbb{R}^M.$$

Theorem (Hohenberg–Kohn)

Suppose that $\psi^{(1)}, \psi^{(2)} \in Q_0$ *are ground states of* $\mathbf{H}(\mathbf{v}^{(1)}, \mathbf{j}^{(1)})$ *and* $\mathbf{H}(\mathbf{v}^{(2)}, \mathbf{j}^{(2)})$ *respectively.*
If $\boldsymbol{\sigma} = \boldsymbol{\sigma}_{\psi^{(1)}} = \boldsymbol{\sigma}_{\psi^{(2)}}$ *and* $\boldsymbol{\xi} = \boldsymbol{\xi}_{\psi^{(1)}} = \boldsymbol{\xi}_{\psi^{(2)}}$*, then* $\psi^{(1)}$ *is also a ground state of* $\mathbf{H}(\mathbf{v}^{(2)}, \mathbf{j}^{(2)})$ *and* $\psi^{(2)}$ *is also a ground state of* $\mathbf{H}(\mathbf{v}^{(1)}, \mathbf{j}^{(1)})$.
Furthermore, $\mathbf{j} = \mathbf{j}^{(1)} = \mathbf{j}^{(2)}$ *and*

- *(Regular case) If* $\boldsymbol{\sigma}$ *is regular, then* $\mathbf{v}^{(1)} = \mathbf{v}^{(2)}$.
- *(Irregular case) Otherwise, for all* $\boldsymbol{\alpha} \in I^{(1)} \cup I^{(2)}$ *there holds*

$$\sum_{n=1}^{N} (\boldsymbol{\sigma}_z^n)_{\boldsymbol{\alpha}\boldsymbol{\alpha}} (v_n^{(1)} - v_n^{(2)}) = E(\mathbf{v}^{(1)}, \mathbf{j}) - E(\mathbf{v}^{(2)}, \mathbf{j}), \tag{10}$$

where $I^{(i)}$ *denotes the set of spinor indices* $\boldsymbol{\alpha}$ *for which* $(\psi^{(i)})^{\boldsymbol{\alpha}} \not\equiv 0$.

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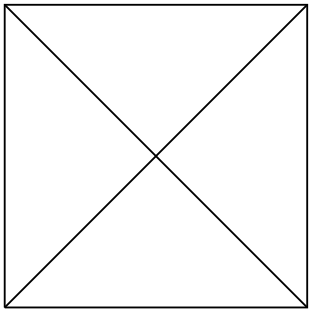
Regular case: Examples $N = 1, 2$

$N = 1$. $\mathcal{R}_1 = (-1, 1)$
$N = 2$. $\mathcal{R}_2 \subset (-1, 1)^2$ is the union of 4 congruent open triangles.



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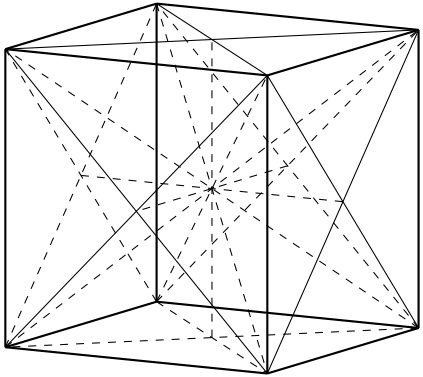
Example $N = 3$

The set $\mathcal{R}_3 \subset (-1, 1)^3$ is the union of 24 congruent open tetrahedra.



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- The regularity property of $\boldsymbol{\sigma}$ can be seen in analogy to finite-lattice DFT [6, Cor. 10].
- Unlike the HK theorem for the electronic Hamiltonian, the potentials are completely determined in the regular case, i.e., not only up to an additive constant.
- The HK itself is nonconstructive, more precisely, it only states the injectivity of the "potential to ground-state density map" $(\mathbf{v}, \mathbf{j}) \mapsto (\boldsymbol{\sigma}, \boldsymbol{\xi})$ and *not* its surjectivity.
- Whenever $(\boldsymbol{\sigma}, \boldsymbol{\xi}) \in [-1, 1]^N \times \mathbb{R}^M$ is the ground-state density of $\mathbf{H}(\mathbf{v}, \mathbf{j})$ for some $(\mathbf{v}, \mathbf{j}) \in \mathbb{R}^N \times \mathbb{R}^M$, we say $(\boldsymbol{\sigma}, \boldsymbol{\xi})$ is v*-representable*.

Levy–Lieb functional

- HK theorem $\implies$ we can formulate the ground-state problem

$$E(\mathbf{v},\mathbf{j}) = \inf_{\substack{\boldsymbol{\psi}\in Q_0 \\ \|\boldsymbol{\psi}\|=1}} \langle \boldsymbol{\psi}, \mathbf{H}(\mathbf{v},\mathbf{j})\boldsymbol{\psi}\rangle \tag{11}$$

  in terms of the density pair $(\boldsymbol{\sigma},\boldsymbol{\xi})$.

- We introduce the constraint manifold that collects all states that map to a given $(\boldsymbol{\sigma},\boldsymbol{\xi}) \in [-1,1]^N \times \mathbb{R}^M$,

$$\mathcal{M}_{\boldsymbol{\sigma},\boldsymbol{\xi}} = \{\boldsymbol{\psi} \in Q_0 : \|\boldsymbol{\psi}\| = 1,\ \boldsymbol{\sigma}_{\boldsymbol{\psi}} = \boldsymbol{\sigma},\ \boldsymbol{\xi}_{\psi} = \boldsymbol{\xi}\}\,. \tag{12}$$

$$
\begin{aligned}
E(\mathbf{v},\mathbf{j}) &= \inf_{\substack{\boldsymbol{\psi}\in Q_0 \\ \|\boldsymbol{\psi}\|=1}} \langle \boldsymbol{\psi}, \mathbf{H}(\mathbf{v},\mathbf{j})\boldsymbol{\psi}\rangle \\
&= \inf_{(\boldsymbol{\sigma},\boldsymbol{\xi})\in[-1,1]^N\times\mathbb{R}^M} \left[\inf_{\boldsymbol{\psi}\in\mathcal{M}_{\boldsymbol{\sigma},\boldsymbol{\xi}}} \langle \boldsymbol{\psi}, \mathbf{H}(\mathbf{v},\mathbf{j})\boldsymbol{\psi}\rangle \right] \\
&= \inf_{(\boldsymbol{\sigma},\boldsymbol{\xi})\in[-1,1]^N\times\mathbb{R}^M} \left[\inf_{\boldsymbol{\psi}\in\mathcal{M}_{\boldsymbol{\sigma},\boldsymbol{\xi}}} \langle \boldsymbol{\psi}, \mathbf{H}_0\boldsymbol{\psi}\rangle + \langle \boldsymbol{\psi}, \mathbf{v}\cdot\boldsymbol{\sigma}_z\boldsymbol{\psi}\rangle + \langle \boldsymbol{\psi}, \mathbf{j}\cdot\mathbf{x}\boldsymbol{\psi}\rangle \right] \\
&= \inf_{(\boldsymbol{\sigma},\boldsymbol{\xi})\in[-1,1]^N\times\mathbb{R}^M} \Big[F_{\mathrm{LL}}(\boldsymbol{\sigma},\boldsymbol{\xi}) + \mathbf{v}\cdot\boldsymbol{\sigma} + \mathbf{j}\cdot\boldsymbol{\xi} \Big]
\end{aligned}
\tag{13}
$$

Levy–Lieb (universal density) functional

Definition

For every $(\boldsymbol{\sigma}, \boldsymbol{\xi}) \in [-1,1]^N \times \mathbb{R}^M$ the *Levy–Lieb* (universal density) *functional* $F_{\mathrm{LL}} : [-1,1]^N \times \mathbb{R}^M \to \mathbb{R}$ is

$$F_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi}) = \inf_{\psi \in \mathcal{M}_{\boldsymbol{\sigma},\boldsymbol{\xi}}} \langle \psi, \mathbf{H}_0 \psi \rangle \tag{14}$$

Immediate question: Is the "$\inf$" in the definition of F_{LL} attained?

Theorem (Existence of an optimizer for F_{LL})

For every $(\boldsymbol{\sigma}, \boldsymbol{\xi}) \in [-1,1]^N \times \mathbb{R}^M$ *there exists a* $\psi \in \mathcal{M}_{\boldsymbol{\sigma},\boldsymbol{\xi}}$ *such that*

$$F_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi}) = \langle \psi, \mathbf{H}_0 \psi \rangle.$$

- Proof is somewhat different from the analogous one in standard DFT [7] and, e.g., generalization to paramagnetic current-DFT [8, 9]:

  there, one exploits the density constraint on the wavefunction to obtain the tightness of the optimizing sequence.
- In our case, the trapping nature of $\mathbf{H}_0$ provides compactness.

Property of F_{LL}

Trial state constructions to derive useful properties of F_{LL}.

Theorem (Displacement rule)

For every $(\boldsymbol{\sigma}, \boldsymbol{\xi}) \in [-1,1]^N \times \mathbb{R}^M$ *the following hold true:*

$$F_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi}) = F_{\mathrm{LL}}(\boldsymbol{\sigma}, 0) + \boldsymbol{\xi} \cdot \boldsymbol{\Lambda}\boldsymbol{\sigma} + |\boldsymbol{\xi}|^2.$$

Or more generaly, for any $\boldsymbol{\zeta} \in \mathbb{R}^M$*,*

$$F_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi} + \boldsymbol{\zeta}) = F_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi}) + 2\boldsymbol{\zeta} \cdot \boldsymbol{\xi} + \boldsymbol{\zeta} \cdot \boldsymbol{\Lambda}\boldsymbol{\sigma} + |\boldsymbol{\zeta}|^2$$

Displacement rule $\implies$ $\boldsymbol{\xi} \mapsto F_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi})$ is smooth and convex for every fixed $\boldsymbol{\sigma} \in [-1,1]^N$.

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Optimizers

Constrained opt: Minimize

$$\langle \boldsymbol{\psi}, \mathbf{H}_0 \boldsymbol{\psi} \rangle \quad \text{s.t.} \quad \boldsymbol{\psi} \in \mathcal{M}_{\boldsymbol{\sigma},\boldsymbol{\xi}} = \{ \boldsymbol{\psi} \in Q_0 : \|\boldsymbol{\psi}\| = 1,\ \boldsymbol{\sigma}_{\boldsymbol{\psi}} = \boldsymbol{\sigma},\ \boldsymbol{\xi}_{\boldsymbol{\psi}} = \boldsymbol{\xi} \}. \tag{15}$$

The tangent space of $\mathcal{M}_{\boldsymbol{\sigma},\boldsymbol{\xi}}$ at $\boldsymbol{\psi} \in \mathcal{M}_{\boldsymbol{\sigma},\boldsymbol{\xi}}$ is given by

$$\mathcal{T}_{\boldsymbol{\psi}}(\mathcal{M}_{\boldsymbol{\sigma},\boldsymbol{\xi}}) = \Big\{ \boldsymbol{\chi} \in Q_0 : \langle \boldsymbol{\psi}, \boldsymbol{\chi} \rangle = 0,\ \langle \boldsymbol{\sigma}_z \boldsymbol{\psi}, \boldsymbol{\chi} \rangle = 0,\ \langle \mathbf{x} \boldsymbol{\psi}, \boldsymbol{\chi} \rangle = 0 \Big\}.$$

Theorem (Optimality)

Let $(\boldsymbol{\sigma}, \boldsymbol{\xi}) \in \mathcal{R}_N \times \mathbb{R}^M$ and suppose that $\psi \in \mathcal{M}_{\boldsymbol{\sigma},\boldsymbol{\xi}}$ is an optimizer of $F_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi})$. Then there exist Lagrange multipliers $E \in \mathbb{R}$, $\mathbf{v} \in \mathbb{R}^N$ and $\mathbf{j} \in \mathbb{R}^M$, such that ψ satisfies the strong Schrödinger equation

$$\mathbf{H}(\mathbf{v}, \mathbf{j})\psi = E\psi \tag{16}$$

and the second-order condition

$$\langle \boldsymbol{\chi}, \mathbf{H}(\mathbf{v}, \mathbf{j})\boldsymbol{\chi} \rangle \geq E\|\boldsymbol{\chi}\|^2, \tag{17}$$

for all $\boldsymbol{\chi} \in \mathcal{T}_{\psi}(\mathcal{M}_{\boldsymbol{\sigma},\boldsymbol{\xi}})$. Moreover,

$$F_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi}) = \langle \psi, \mathbf{H}_0\psi \rangle = E - \mathbf{v} \cdot \boldsymbol{\sigma} - \mathbf{j} \cdot \boldsymbol{\xi}. \tag{18}$$

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The second-order information (17) about a minimizer gives a result which is analogous to the Aufbau principle in Hartree–Fock theory.

Theorem (Optimizers are low-lying eigenstates)

Let $(\boldsymbol{\sigma}, \boldsymbol{\xi}) \in \mathcal{R}_N \times \mathbb{R}^M$, and suppose that $\psi \in \mathcal{M}_{\boldsymbol{\sigma},\boldsymbol{\xi}}$ is an optimizer of $F_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi})$, with Lagrange multipliers $E \in \mathbb{R}$, $\mathbf{v} \in \mathbb{R}^N$ and $\mathbf{j} \in \mathbb{R}^M$, so that (16) *and* (17) *holds true. Then ψ is at most the $(N+M)$th excited eigenstate of $\mathbf{H}(\mathbf{v}, \mathbf{j})$.*

Any $(\boldsymbol{\sigma}, \boldsymbol{\xi}) \in \mathcal{R}_N \times \mathbb{R}^M$, while not proven to be pure-state v-representable in the usual sense, can be called “low-lying excited-pure-state v-representable”.

The Universal Density-Functional $M = N = 1$

$$F^{\lambda}_{\mathrm{LL}}(\sigma,\xi) = \min_{\psi\in\mathcal{M}_{\sigma,\xi}} \langle \psi, H^{\lambda}_0 \psi\rangle \qquad F^{\lambda}_{\mathrm{L}}(\sigma,\xi) = \max_{(v,j)} (E^{\lambda}(v,j) - v\sigma - j\xi)$$

$$H^{\lambda}_0 = (-\Delta + |x|^2)\mathbb{1}_{\mathbb{C}^2} + \lambda x \sigma_z - t\sigma_x$$

Corollary ($M = N = 1$)

Consider a regular density pair $(\sigma,\xi) \in (-1,1)\times\mathbb{R}$. Then the following holds:

(i) *(v-representability) The (σ,ξ) is uniquely pure-state v-representable.*

(ii) *(equivalence of functionals) $F^{\lambda}_{\mathrm{LL}}(\sigma,\xi) = F^{\lambda}_{\mathrm{L}}(\sigma,\xi)$.*

(iii) *(differentiability) The $F^{\lambda}_{\mathrm{LL}}$ is differentiable at (σ,ξ) and $(v,j) = -\nabla F^{\lambda}_{\mathrm{LL}}(\sigma,\xi)$ is its representing external potential pair.*

Addiabatic connection formula

$$F^{\lambda}_{\mathrm{LL}}(\sigma,\xi) = F^{0}_{\mathrm{LL}}(\sigma,\xi) + \int_0^{\lambda} \langle \psi^{\nu}, x\sigma_z \psi^{\nu} \rangle \, d\nu$$

where

$$\psi^{\nu} = \psi^{\nu}(\sigma,\xi) = \operatorname*{argmin}_{\psi \in \mathcal{M}_{\sigma,\xi}} \langle \psi, H^{\nu}_0 \psi \rangle$$

$$F^{0}_{\mathrm{LL}}(\sigma,\xi) = 1 - t\sqrt{1-\sigma^2} + \xi^2$$

In addition,

$$\int_0^{\lambda} \langle \psi^{\nu}, x\sigma_z \psi^{\nu} \rangle \, d\nu = \lambda\sigma\xi - \frac{1}{4}\lambda^2(1-\sigma^2) + I^{\lambda}(\sigma),$$

$$I^{\lambda}(\sigma) \sim -\int_0^{\lambda} \int_{\mathbb{R}} (\psi(\sigma,0)^{\nu})'_{+} (\psi(\sigma,0)^{\nu})_{-} \, dx d\nu$$

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Thank you for your attention!

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References I

1. **Lammert, P. E.** Differentiability of Lieb functional in electronic density functional theory. *Int. J. Quantum Chem.* **107,** 1943–1953 (10 2007).
2. **Kvaal, S., Ekström, U., Teale, A. M. & Helgaker, T.** Differentiable but exact formulation of density-functional theory. *J. Chem. Phys.* **140,** 18A518. https://doi.org/abs/10.1063/1.4867005 (2014).
3. **Penz, M., Csirik, M. A. & Laestadius, A.** Density-potential inversion from Moreau–Yosida regularization. *Electron. Struct.* **5,** 014009 (2023).
4. **Herbst *et al.*, M. F.** Kohn-Sham inversion with mathematical guarantees. *Phys. Rev. B* **111,** 205143 (2025).
5. **Kim, M.-C., Sim, E. & Burke, K.** Understanding and Reducing Errors in Density Functional Calculations. *Phys. Rev. Lett.* **111,** 073003. https://link.aps.org/doi/10.1103/PhysRevLett.111.073003 (7 Aug. 2013).

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References II

6. **Penz, M. & van Leeuwen, R.** Density-functional theory on graphs. *J. Chem. Phys.* **155** (2021).
7. **Lieb, E. H.** Density Functionals for Coulomb-Systems. *Int. J. Quantum Chem.* **24,** 243–277. https://doi.org/10.1002/qua.560240302 (1983).
8. **Laestadius, A.** Density functionals in the presence of magnetic field. *Int. J. Quantum Chem.* **114,** 1445–1456 (2014).
9. **Kvaal, S., Laestadius, A., Tellgren, E. & Helgaker, T.** Lower Semicontinuity of the Universal Functional in Paramagnetic Current–Density Functional Theory. *J. Phys. Chem. Lett.* **12,** 1421–1425 (2021).

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