IWOTA 2023

The unique-continuation property in density-functional theory

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Acknowledgements

Funding:

ERC StG project REGAL (No. 101041487)

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European Research Council

Established by the European Commission

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- N electrons, positions $r_i \in \mathbb{R}^3$ $(1 \leq i \leq N)$
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- N-body Hamiltonian

$$H = H^N = \underbrace{\sum_{i=1}^{N} t_{r_i}}_{T} + \underbrace{\frac{1}{2} \sum_{1 \leq i \neq j \leq N} w(r_i - r_j)}_{W} + \underbrace{\sum_{i=1}^{N} v(r_i)}_{V} \tag{1}$$

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UCP (from sets of pos. measure)

If $(H - E)\psi = 0$ and $\psi = 0$ on a set of pos. measure, is $\psi = 0$ a.e.?

Example (Coulomb systems)

Nuclei with charges $\{Z_j\}_{j=1}^M$ and positions $\{R_j\}_{j=1}^M$:

$$V = \sum_{i=1}^N v(r_i) = -\sum_{i=1}^N \sum_{j=1}^M Z_j |r_i - R_j|^{-1} \tag{2}$$

Two-electron interaction

$$W = \sum_{i<j} w(r_i - r_j) = \sum_{i<j} |r_i - r_j|^{-1} \tag{3}$$

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Kinetic operator

Canonical: $t_i = -\Delta_i$
Physical: $t_i = (\mathrm{i}\nabla_i + \vec{A}(r_i))^2$, magnetic field $\vec{B} = \nabla \times \vec{A}$

- *N*-body Hamiltonian

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- Here $d = 3N$. Typical results have requirement on U that depends on the particle number
- In magnetic fields, this also applies to $\vec{A}$

Weak UCP of Georgescu (1980), Schechter–Simon (1980)

Assumptions on the potentials in $H = H^N$ independent of N that gives weak UCP (ψ vanishes on an open set $\implies \psi = 0$).

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UCP of L., Penz & Benedicks (2020)

UCP for $H = H^N$ based on Kurata (1997) and Regbaoui (2001). Conditions in terms of the Kato class, and extra assumption on the "virial term"

$$2U(r_1, \dots, r_N) + (r_1, \dots, r_N) \cdot \nabla_{\mathbb{R}^{3N}} U(r_1, \dots, r_N)$$

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UCP of Garrigue (2018)

UCP for $H = H^N$ for $v, w \in L^p_{\mathrm{loc}}(\mathbb{R}^3)$, $p > 2$.

Density-functional theory

- $H\psi = E\psi, \qquad E = E(v)$ ground-state energy

- $E(v) = \inf\{\langle\psi, H\psi\rangle : \underbrace{\psi \in H^1(\mathbb{R}^{3N}), \|\psi\| = 1}_{=:\mathcal{W}_N}\}$

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One-body particle density

$$\rho_\psi(r) = N \int_{\mathbb{R}^{3(N-1)}} |\psi|^2, \tag{5}$$

$\psi \in \mathcal{W}_N \implies \rho_\psi \in L^1 \cap L^3$ (Lieb, 1983)

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Hohenberg–Kohn (1964)

The particle density ρ determines a quantum mechanical system of N electrons completely.

Structure of H,

$$H = \underbrace{T + W}_{=:H_0} + V = H_0 + \sum_{i=1}^{N} v(r_i)$$

$$\langle \psi, H\psi \rangle = \langle \psi, (H_0 + V)\psi \rangle = \underbrace{\langle \psi, H_0\psi \rangle}_{\text{universal}} + \int_{\mathbb{R}^3} v\rho_\psi$$

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Ground-state energy via "constrained-search" (Levy, 1979)

$$E(v) = \inf_{\psi \in \mathcal{W}_N} \left\{ \langle \psi, H_0\psi \rangle + \int v\rho_\psi \right\}$$
$$= \inf_{\rho} \left\{ \underbrace{\inf\{\langle \psi, H_0\psi \rangle : \psi \in \mathcal{W}_N, \rho_\psi = \rho\}}_{\tilde{F}(\rho)} + \int v\rho \right\}$$

- Levy–Lieb functional $\tilde{F}$

$$\tilde{F}(\rho) = \inf_{\psi \in \mathcal{W}_N : \rho_\psi = \rho} \{\langle \psi, H_0 \psi \rangle\}$$

- Ground-state energy

$$E(v) = \inf_{\rho} \left\{ \tilde{F}(\rho) + \int v\rho \right\} \tag{6}$$

- Lieb functional

$$F(\rho) = \sup_{v} \left\{ E(v) - \int v\rho \right\} \tag{7}$$

Fenchel–Young (FY) inequality

Eqs. (6) and (7) different ways to satuarate FY inequality

$$E(v) - F(\rho) \leq \int v\rho$$

- ρ_{gs} ground-state density
- The naive Euler–Lagrange

$$\frac{\delta}{\delta\rho}F(\rho_{\mathrm{gs}}) + v = \mu \tag{8}$$

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Non-differentiability

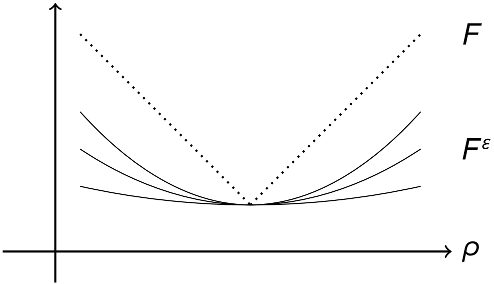
$\frac{\delta}{\delta\rho}F$ is **NOT** available since F is **NOT** differentiable (Lammert, 2007)

Hohenberg–Kohn variational principle

$E(v) = \min_\rho \{F(\rho) + \int v\rho\}$ $F(\rho)$ is highly irregular

Moreau–Yosida regularization

$E^\varepsilon(v) = \min_\rho \{F^\varepsilon(\rho) + \int v\rho\}$ $F^\varepsilon(\rho)$ is differentiable



Moreau–Yosida regularization

- X reflexive, strictly (uniform) convex
- Convex, lower semicontinuous functional $f : X \to \cup\{+\infty\}$
- MY regularization

$$f^\varepsilon(\rho) = \inf_{\rho' \in X} \left\{ f(\rho') + \tfrac{1}{2\varepsilon} \|\rho - \rho'\|_X^2 \right\}, \quad \varepsilon > 0 \tag{9}$$

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- Infimum is attained at a unique point:
  proximal mapping $\Pi_f^\varepsilon : X \to X$

$$\rho^\varepsilon := \Pi_f^\varepsilon(\rho) = \mathrm{argmin}_{\rho' \in X} \left\{ f(\rho') + \tfrac{1}{2\varepsilon} \|\rho - \rho'\|_X^2 \right\}. \tag{10}$$

- Suppose $\rho \in X, \quad v \in X^*$ (e.g. $X = L^3(\mathbb{R}^3)$)
- Let J be the duality mapping from X to X^*
- Derivative

$$\frac{\delta}{\delta\rho} F^\varepsilon(\rho) = \frac{1}{\varepsilon} J(\rho - \Pi_F^\varepsilon(\rho))$$

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$$\frac{\delta}{\delta\rho} F^\varepsilon(\rho) = \frac{1}{\varepsilon} J(\rho - \Pi_F^\varepsilon(\rho))$$

- Euler Lagrange

$$\frac{\delta}{\delta\rho} F^\varepsilon(\rho) + v = 0$$

Thus

$$v = -\frac{1}{\varepsilon} J(\rho - \rho^\varepsilon) \tag{11}$$

- Eq. (11) has connection to what practitioners do (Penz et. al, 2023)

Hohenberg–Kohn theorem and UCP

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Example ($N = 1$)

Let $\rho(r) = C_\gamma e^{-2\gamma|r|} > 0$, then

$$v(r) = -\frac{\gamma^2}{|r|} + \text{constant} \tag{12}$$

Follows from Schrödinger equation

$$(-\Delta + v)\sqrt{\rho} = \text{constant}\sqrt{\rho}$$

Theorem (HK1, Penz et al. (2023))

Let ψ_k be a ground state of $H = H(v_k)$, $k = 1, 2$. If $\psi_1, \psi_2 \mapsto \rho$, then ψ_1 is also a ground state of $H(v_2)$ and ψ_2 is also a ground state $H(v_1)$.

Proof.

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Proof.

- $$E(v_k) = \underbrace{\min_{\psi \mapsto \rho} \langle \psi, H_0 \psi \rangle}_{(*)} + \int v_k \rho \tag{13}$$
- $(*)$ in Eq. (13) completely determined by the fixed ρ
- The density alone already defines the ground state, irrespective of the potential v_1 or v_2.

Theorem (HK2, Penz et al. (2023))

If two potentials share any common eigenstate and if that eigenstate is non-zero almost everywhere, then the potentials are equal up to a constant.

Proof.

- If v_1, v_2 share a common eigenstate ψ, it holds

$$(H_0 + \sum_i v_1(r_i))\psi = E(v_1)\psi,$$
$$(H_0 + \sum_i v_2(r_i))\psi = E(v_2)\psi,$$

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$$(v_1(r_1) - v_2(r_1))\psi = (E(v_1) - E(v_2))\psi - \sum_{i=2}^{N}(v_1(r_i) - v_2(r_i))\psi \qquad (14)$$

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- $\psi \neq 0$ a.e. $\implies$ $v_1(r_1) - v_2(r_1) = \text{constant}$

Theorem (Garrigue's UCP (2018))

Suppose $v, w \in L^p_{\mathrm{loc}}(\mathbb{R}^3)$ with $p > 2$. If a solution ψ to the Schrödinger equation vanishes on a set of positive measure, then $\psi = 0$.

Idea of proof.

- Recall $U = V + W = \sum_i v(r_i) + \sum_{i<j} w(r_i - r_j)$,

$$|U|^2 1_{B_R} \leq \varepsilon_N(-\Delta)^{3/2} + c_R, \quad B_R \subset \mathbb{R}^{3N} \implies -\Delta + U \quad \text{has the UCP} \quad (15)$$

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- Carleman inequality (Tataru 2004): $s \in [0, 2]$, $\tau > \tau_0$,

$$\tau^{\frac{3}{2}-s}\|(-\Delta + \tau^2)^{s/2} e^{\tau\varphi} f\|_{L^2} \leq \kappa_N \|e^{\tau\varphi}\|_{L^2(B_1)}, \quad f \in H^2_0(B_1) \subset H^2(\mathbb{R}^{3N}) \tag{16}$$

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$e^{\varphi} + |\nabla\varphi| \leq C|r|^{-1}$, $|\Delta\varphi| \leq C|r|^{-2}$

Corollary (HK, originally by Hohenberg & Kohn (1964))

Let $p > 2$. Suppose the class of potentials is $L^p(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$. If two potentials share a common ground-state density, then they are equal up to a constant.

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Lieb's formulation of DFT

HK is not yet proven for $v \in L^{3/2}(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$.

Thank you for your attention!