Mathematical ideas for DFT

Mihály A. Csirik

Department of Computer Science, Oslo Metropolitan University

Oslo
2024

Rocking the boat

"The theoretical physicist typically wants to get an answer. It's like going over stepping stones. He can jump when he feels like jumping as long as he gets to the other side.

But in mathematical physics you don't do that.

If the final mathematical result is going to be effective, it's got to be completely rigorous." — E. H. Lieb

"They muddy the water, to make it seem deep." — Nietzsche

Overview

Disclaimer: Some ideas are very much speculative and work-in-progress.

Contents:

- **Known results**
- **Motivation I.:** "exotic" DFTs
  1. Non-collinear spin-polarized DFT
  2. Quantum-electrodynamical DFT for the Pauli–Fierz model
- ~~**Motivation II.:** (meta-) (generalized-) gradient expansion/approximation, etc.~~
  1. Separation of center-of-mass and internal coordinates $\Longrightarrow$
     1. A rigorous treatment of the non-uniform electron gas
- **Proposal: "dressed DFT"**
  1. Instead of the scalar density, **matrix-valued** "dressed" densities.
  2. A series of complications due to topological obstructions to global diagonalization of matrix-valued functions.

Known results for ordinary DFT

Notations I.

- In the first half we consider the grand-canonical functionals for technical reasons.
- $\Gamma = \Gamma_0 \oplus \Gamma_1 \oplus \ldots$ denotes a fermionic Fock space state that commutes with the number operator. The set of such states with finite kinetic energy is denoted by $\mathcal{D}$.
- N-representability set:

$$\mathcal{I} = \{\rho \in L^1(\mathbb{R}^3, \mathbb{R}_+) : \boldsymbol{\nabla}\sqrt{\rho} \in L^2(\mathbb{R}^3)\}$$

- **Grand-canonical Levy–Lieb functional:**

$$\boxed{F(\rho) = \inf_{\substack{\Gamma \in \mathcal{D} \\ \rho_\Gamma = \rho}} \sum_{n \geq 1} \mathrm{Tr}\left(-\sum_{1 \leq j \leq n} \boldsymbol{\Delta}_{x_j} + \sum_{1 \leq j < k \leq n} \frac{1}{|x_j - x_k|} \right) \Gamma_n}$$

- **Direct (classical) Coulomb energy:**

$$\boxed{D(\rho) = \frac{1}{2} \iint_{\mathbb{R}^3 \times \mathbb{R}^3} \frac{\rho(x)\rho(x')}{|x - x'|} \, \mathrm{d}x \mathrm{d}x'}$$

Notations II.

- **Indirect energy:**

$$\boxed{E(\rho) = F(\rho) - D(\rho)}$$

Has a sign, but bounded from below by the Lieb–Thirring-, and the Lieb–Oxford inequalities.

- **Exchange–correlation energy:**

$$\boxed{E_{\mathrm{xc}}(\rho) = F(\rho) - (T(\rho) + D(\rho))}$$

Nonpositive and similarly bounded from below.

The '83 Lieb article

- The first foundation of the mathematically rigorous study of DFT.[1]
- Considers the canonical case only, but easily extends to the grand-canonical one.
- **Main points:**
  1. The N-representability set is easliy described, the v-representability set is complicated.
  2. Hohenberg–Kohn-type functionals are not well-defined.
  3. In the Levy–Lieb (i.e. pure state constraint search) functional the infimum is attained. But Levy–Lieb type functionals are **nonconvex**.
  4. The Lieb functional, which is the Legendre–Fenchel conjugate of the ground-state energy $v \mapsto -E_0(-v)$ **is** convex and l.s.c. Can be given as the convex envelope of the Levy–Lieb functional. The infimum is also attained in the Lieb functional.
- **Later points:**
  1. The universal density functionals are nowhere differentiable.[2]
  2. Moreau–Yosida regularization can be used to make them differentiable.[3]
  3. Etc.

---

[1] E. H. Lieb. *Density functionals for Coulomb systems*, 1983.

[2] P. E. Lammert. *Differentiability of Lieb functional in electronic density functional theory*, 2007.

[3] S. Kvaal et al. *Differentiable but exact formulation of density-functional theory*, 2014.

Thermodynamic limit for the UEG I.

- Lewin, Lieb and Seiringer defined the **uniform electron gas (UEG)** using the grand-canonical Levy–Lieb functional.[4]
- $\implies$ **An important theoretical application of DFT!**
- Let $0 \leq \eta \in C_c^\infty(\mathbb{R}^3)$ be such that $\operatorname{supp} \eta \subset B_1$, $\int_{\mathbb{R}^3} x\eta(x)\,\mathrm{d}x = 0$ and $\int_{\mathbb{R}^3} \eta = 1$ (**regularization function**)
- For any bounded domain $\Omega \subset \mathbb{R}^3$ define the **indirect energy per volume** at constant density $\rho_0 \in \mathbb{R}_+$,

$$\boxed{e_{\Omega,\eta}(\rho_0) = \frac{1}{|\Omega|} E((\mathbb{1}_\Omega * \eta)\rho_0)}$$

- We can similarly define the **exchange-correlation energy per volume** $e^{\mathrm{xc}}_{\Omega,\eta}(\rho_0)$ and **kinetic energy per volume** $\tau_{\Omega,\eta}(\rho_0)$.
- We have a bound

$$\boxed{-c_{\mathrm{LO}}\rho_0^{4/3} \leq e_{\mathrm{UEG}}(\rho_0) \leq c_{\mathrm{TF}}\rho_0^{5/3}}$$

[4] M. Lewin et al. *Statistical mechanics of the uniform electron gas*, 2018.

Thermodynamic limit for the UEG II.

- A sequence of bounded domains $\{\Omega_N\}_{N\in\mathbb{N}} \subset \mathbb{R}^3$ has uniformly regular boundary (a.k.a. Fischer-regular boundary) if $|\partial\Omega_N + B_r| \leq Cr|\Omega_N|^{2/3}$ for all $r \leq \frac{|\Omega_N|^{2/3}}{C}$
- Let $\eta_\delta = \delta^{-3}\eta(\delta^{-1}\cdot)$ for $\delta > 0$.

Theorem (LLS18, Uniform electron gas)

Let $\rho_0 \in \mathbb{R}_+$ and let $\{\Omega_N\}_{N\in\mathbb{N}} \subset \mathbb{R}^3$ be a sequence of bounded domains such that $|\Omega_N| \to \infty$ it has a uniformly regular boundary. Let $\delta_N > 0$ be any sequence such that $\delta_N/|\Omega_N|^{1/3} \to 0$ and $\delta_N|\Omega_N|^{1/3} \to \infty$. Then the following thermodynamic limit exists

$$\boxed{e_{\mathrm{UEG}}(\rho_0) = \lim_{N\to\infty} e_{\Omega_N,\eta_{\delta_N}}(\rho_0)}$$

and is independent of the choice of the sequences $\{\Omega_N\}$, $\{\delta_N\}$ and η.

- The spin-polarized generalization is trivial.
- We generalized this to the magnetic case, where the vorticity is also held fixed[5]
- **Research idea:** periodic boundary conditions.

[5] M. A. Cs., A. Laestadius, E. I. Tellgren. *Thermodynamic limit for the magnetic uniform electron gas and representability of density-current pairs*, 2024.

Error estimate for LDA I.

- Using the above definition of the UEG, Lewin, Lieb and Seiringer went on to analyze the **local density approximation (LDA)** in DFT.[6]

Theorem (LLS19, Local density approximation)

Let $p > 3$ *and* $0 < \theta < 1$ *such that* $2 \leq p\theta \leq 1 + p/2$. *There exists a constant* $C = C(p, \theta) > 0$ *such that for all* $\varepsilon > 0$ *and* $0 \leq \rho \in L^1(\mathbb{R}^3) \cap L^2(\mathbb{R}^3)$ *with* $\nabla\sqrt{\rho} \in L^2(\mathbb{R}^3)$ *and* $\nabla\rho^\theta \in L^p(\mathbb{R}^3)$ *the bound*

$$\left| E(\rho) - \int_{\mathbb{R}^3} e_{\mathrm{UEG}}(\rho(x))\,\mathrm{d}x \right| \leq \varepsilon \int_{\mathbb{R}^3} (\rho + \rho^2) + \frac{C(1+\varepsilon)}{\varepsilon} \int_{\mathbb{R}^3} |\nabla\sqrt{\rho}|^2 + \frac{C}{\varepsilon^{4p-1}} \int_{\mathbb{R}^3} |\nabla\rho^\theta|^p$$

holds true.

- Difficult and long proof.
- **Poses an important challenge:** Is there an analogous result for gradient approximation?
- Trivial to generalize to collinear spin DFT. **Research idea:** noncollinear spin DFT.

[6] M. Lewin et al. *The local density approximation in density functional theory*, 2019.

Error estimate for LDA II.

- **Ingredients of the proof:**
  1. Spatial decoupling upper bound using the tensor product of disjointly supported states and a clever partition of unity with holes.
  2. Spatial decoupling lower bound using the IMS localization formula and the Graf–Schenker inequality.
  3. Lipschitz regularity of the function $e_{\mathrm{UEG}}(\rho_0)$.
  4. Local replacement of the density with a constant on **big** boxes.
  5. This in turn is based on the following corollary of the **convexity** of $F(\rho)$ combined with a kinetic energy estimate:

Theorem (Rough subadditivity estimate)

Let $\rho_1, \rho_2 \in \mathcal{I}$, then for all $0 < \varepsilon \leq 1$ there holds

$$E(\rho_1+\rho_2) \leq E(\rho_1) + C\varepsilon \int_{\mathbb{R}^3} (\rho_1^{5/3} + \rho_1^{4/3}) + C\varepsilon^{-2/3} \int_{\mathbb{R}^3} \rho_2^{5/3} + C \int_{\mathbb{R}^3} |\nabla \sqrt{\rho_2 + \varepsilon\rho_1}|^2 + \frac{1-\varepsilon}{\varepsilon} D(\rho_2)$$

This is then used with $\rho_1 = \underline{\rho}$ and $\rho_2 = \rho - \underline{\rho}$ (...)

“Exotic” DFTs

Pauli Hamiltonian

$$H^{v,\mathbf{B}} = \sum_{1\le n\le N} \Big[-\mathbf{\Delta}_{x_n} + \boldsymbol{\sigma}_n \cdot \mathbf{B}(x_n) + v(x_n) \Big] + \sum_{1\le n<m\le N} \frac{1}{|x_n - x_m|}$$

on the Hilbert space $L_a^2((\mathbb{R}^3 \times \{\uparrow,\downarrow\})^N) \simeq L_a^2(\mathbb{R}^{3N}, \mathbb{C}^{2^N})$

- Here σ_n is the vector of usual Pauli matrices lifted to the N-particle space:

$$\sigma_n^j = \mathbb{1} \otimes \ldots \otimes \mathbb{1} \otimes \underbrace{\sigma^j}_{n\text{th}} \otimes \mathbb{1} \otimes \ldots \otimes \mathbb{1} \quad (n = 1, \ldots, N), \quad \sigma^1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma^2 = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \sigma^3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

- **Densities:**
  1. Either the matrix of spin-polarized densities $\boldsymbol{\rho}(x) = \rho^{\alpha\beta}(x)$ **or equivalently**
  2. The total electron density $\rho(x) = \rho^{\uparrow\uparrow}(x) + \rho^{\downarrow\downarrow}(x)$, and the magnetization $\mathbf{m}(x)$ given by $\mathbf{m} = \mathrm{Tr}_{\mathbb{C}^2}\, \boldsymbol{\sigma}\boldsymbol{\rho}$, so that $\boldsymbol{\rho} = \frac{1}{2}\rho\mathbb{1} + \mathbf{m} \cdot \boldsymbol{\sigma}$
- We have $\langle \Psi, H^{v,\mathbf{B}}\Psi \rangle = \langle \Psi, H^{0,0}\Psi \rangle + \langle v, \rho_\Psi \rangle + \langle \mathbf{B}, \mathbf{m}_\Psi \rangle$
- Only **partial** Hohenberg–Kohn result[7], but the theory still makes sense.
- N-representability is proved[8]

[7] L. Garrigue. *Hohenberg-Kohn theorems for interactions, spin and temperature*, 2019.

[8] D. Gontier. *N-Representability in Noncollinear Spin-Polarized Density-Functional Theory*, 2013

$$H^{v,\mathbf{A}} = \sum_{1\le n\le N} \Big[\big(-i\boldsymbol{\nabla}_{x_n} + \mathbf{A}(x_n)\big)^2 + v(x_n)\Big] + \sum_{1\le n<m\le N} \frac{1}{|x_n - x_m|}$$

- **Densities:**
  1. The total electron density $\rho(x)$
  2. The paramagnetic current density $\mathbf{j}^{\mathrm{p}}(x)$
- We have $\langle \Psi, H^{v,\mathbf{A}}\Psi\rangle = \langle \Psi, H^{0,0}\Psi\rangle + \langle v + |\mathbf{A}|^2, \rho_\Psi\rangle + 2\langle \mathbf{A}, \mathbf{j}^{\mathrm{p}}_\Psi\rangle$
- **No** Hohenberg–Kohn theorem[9]
- However, the Vignale–Rasolt functional

$$F_{\mathrm{VR}}(\rho, \mathbf{j}^{\mathrm{p}}) = \inf_{\substack{\|\Psi\|=1\\ \rho_\Psi=\rho\\ \mathbf{j}^{\mathrm{p}}_\Psi=\mathbf{j}^{\mathrm{p}}}} \langle \Psi, H^{0,0}\Psi\rangle$$

is well-behaved[10] [11]

[9] A. Laestadius, M. Benedicks. *HK theorems in the presence of magnetic field*, 2014.

[10] A. Laestadius. *Density functionals in the presence of magnetic field*, 2014

[11] S. Kvaal et al. *Lower Semicontinuity of the Universal Functional in Paramagnetic Current-Density Functional Theory*, 2021.

Pauli–Fierz Hamiltonian:

$$H^{v,\mathbf{A}} = \frac{1}{2}\sum_{1\le n\le N}\sum_{1\le j\le 3}\left[\sigma_n^j \otimes \left(-\partial_{x_n^j} + A^j(x_n)\right)\otimes \mathbb{1} - e\sigma_n^j \otimes \mathfrak{A}^j(x_n)\right]^2$$

$$+\,\mathbb{1}\otimes \sum_{1\le n\le N} v(x_n)\otimes \mathbb{1} + \mathbb{1}\otimes \sum_{1\le n<m\le N}\frac{1}{|x_n - x_m|}\otimes \mathbb{1} + \mathbb{1}\otimes \mathbb{1}\otimes H_{\mathrm{rad}}$$

- N-electron Hilbert space: $\mathfrak{H}_a^N = L_a^2((\mathbb{R}^3\times\{\uparrow,\downarrow\})^N) \simeq \mathbb{C}^{2^N}\otimes L_a^2(\mathbb{R}^{3N})$
- One-photon Hilbert space: $\mathfrak{H}_{\mathrm{ph}} := L^2(\mathbb{R}^3\times\{1,2\}) \simeq L^2(\mathbb{R}^3,\mathbb{C}^2)$
- Photon Fock space: $\mathcal{F}_{\mathrm{ph}} := \mathcal{F}_s(\mathfrak{H}_{\mathrm{ph}})$
- The total Hilbert space is $\mathfrak{H}_a^N\otimes\mathcal{F}_{\mathrm{ph}} \simeq L_a^2((\mathbb{R}^3\times\{\uparrow,\downarrow\})^N,\mathcal{F}_{\mathrm{ph}})$
- Fock representation of the **CCR**:

$$[a(k,\nu), a^\dagger(k',\nu')] = \delta(k-k')\delta_{\nu,\nu'}$$

$$[a^\dagger(k,\nu), a^\dagger(k',\nu')] = [a(k,\nu), a(k',\nu')] = 0$$

- The **UV cutoff vector potential operator** is a multiplication operator on $L^2(\mathbb{R}^{3N}, \mathcal{F}_{\text{ph}})$,

$$\mathfrak{A}^j(x) = \sum_{\nu=1,2} \int_{|k|\leq\Lambda} \frac{\varepsilon_\nu^j(k)}{\sqrt{|k|}} \left[e^{-ik\cdot x} a^\dagger(k,\nu) + e^{ik\cdot x} a(k,\nu) \right] \mathrm{d}k$$

- Here for $\nu = 1, 2$, $\boldsymbol{\varepsilon}_\nu : \mathbb{R}^3 \to \mathbb{R}^3$ are (a.e.) orthonormal **polarization vectors** such that $k \cdot \boldsymbol{\varepsilon}_\nu(k) = 0$ for a.a. $k \in \mathbb{R}^3$.
- The projector onto the plane of polarization,

$$\sum_{\nu=1,2} \boldsymbol{\varepsilon}_\nu(k) \otimes \boldsymbol{\varepsilon}_\nu(k) = \mathbb{1} - \frac{k \otimes k}{|k|^2}$$

is an observable quantity (i.e. k), but the vectors $\boldsymbol{\varepsilon}_\nu(k)$ are not.
- The **free field Hamiltonian** H_{rad} is a *nonlocal* operator in position space:

$$H_{\text{rad}} = \sum_{\nu=1,2} \int_{\mathbb{R}^3} |k|\, a^\dagger(k,\nu) a(k,\nu)\, \mathrm{d}k = 4\pi \sum_{\nu,\nu'=1,2} \iint_{\mathbb{R}^3\times\mathbb{R}^3} \frac{\|a(x,\nu)\Psi - a(x',\nu')\Psi\|^2}{|x-x'|^4} \mathrm{d}x \mathrm{d}x'$$

Quantum-electrodynamical DFT III.

- **For simplicity we disregard spin:**

$$H^{v,\mathbf{A}} = \frac{1}{2}\sum_{1\le n\le N}\Big[(-\boldsymbol{\nabla}_{x_n} + \mathbf{A}(x_n))\otimes\mathbb{1} - e\mathfrak{A}(x_n)\Big]^2$$
$$+ \sum_{1\le n\le N} v(x_n)\otimes\mathbb{1} + \sum_{1\le n<m\le N}\frac{1}{|x_n - x_m|}\otimes\mathbb{1} + \mathbb{1}\otimes H_{\mathrm{rad}}$$

- We may write

$$\frac{1}{2}(-i\boldsymbol{\nabla}_x + \mathbf{A}(x) - e\mathfrak{A}(x))^2$$
$$= \frac{1}{2}\big(-i\boldsymbol{\nabla}_x - e\mathfrak{A}(x)\big)^2 + 2\mathbf{A}(x)\cdot(-i\boldsymbol{\nabla}_x - e\mathfrak{A}(x)) + \frac{1}{2}|\mathbf{A}(x)|^2$$

Taking expectation values at $\Psi \in \mathfrak{H}_a^N \otimes \mathcal{F}_{\mathrm{ph}}$, we obtain for the mixed terms

$$2\langle\Psi, \mathbf{A}(x_1)\cdot(-i\boldsymbol{\nabla}_{x_1} - e\mathfrak{A}(x_1))\Psi\rangle$$
$$= \int_{\mathbb{R}^3}\mathbf{A}(x_1)\cdot 2\,\mathrm{Re}\sum_{M=0}^{\infty}\iint_{(\mathbb{R}^3\times\{1,2\})^M\times\mathbb{R}^{3(N-1)}}\overline{\psi^M(X;W)}(i\boldsymbol{\nabla}_{x_1} - e\mathfrak{A}(x_1))\psi^M(X;W)\,\mathrm{d}W\,\mathrm{d}X$$

Quantum-electrodynamical DFT IV.

- We recognize the paramagnetic current density,

$$\mathbf{j}^{\mathrm{p}}_{\Psi}(x_1) = 2N\,\mathrm{Im}\langle\Psi, \boldsymbol{\nabla}_{x_1}\Psi\rangle_{x_1},$$

  and call the quantity

$$\mathbf{j}^{\mathrm{ph}}_{\Psi}(x_1) = 2N\langle\Psi, \mathfrak{A}(x_1)\Psi\rangle_{x_1}.$$

  the radiation field-induced current.

- In summary, we obtained

$$\boxed{\langle\Psi, H^{v,\mathbf{A}}\Psi\rangle = \langle\Psi, H^{0,0}\Psi\rangle + \langle v + \tfrac{1}{2}|\mathbf{A}|^2, \rho_\Psi\rangle + \langle\mathbf{A}, \mathbf{j}^{\mathrm{p}}_{\Psi}\rangle - e\langle\mathbf{A}, \mathbf{j}^{\mathrm{ph}}_{\Psi}\rangle}$$

- So actually $\mathbf{A}$ couples to the "combined current" $\mathbf{j}^{\mathrm{c}}_{\Psi} = \mathbf{j}^{\mathrm{p}}_{\Psi} - e\mathbf{j}^{\mathrm{ph}}_{\Psi}$.
- This motivates to define the functional

$$F^{e,\Lambda}(\rho, \mathbf{j}^{\mathrm{c}}) = \inf_{\substack{\Psi\in\mathfrak{H}^N_a\otimes\mathcal{F}_{\mathrm{ph}}, \|\Psi\|=1 \\ \rho_\Psi = \rho \\ \mathbf{j}^{\mathrm{c}}_{\Psi} = \mathbf{j}^{\mathrm{c}}}} \langle\Psi, H^{0,0}\Psi\rangle$$

Quantum-electrodynamical DFT VI.

- Let us consider the **zero coupling limit** $e \to 0$.
- Formally putting $e = 0$, we see that we **recover the Vignale–Rasolt functional:**

$$F^{0,\Lambda}(\rho, \mathbf{j}^{\mathrm{p}}) = F_{\mathrm{VR}}(\rho, \mathbf{j}^{\mathrm{p}})$$

In fact, the constraint reads $\mathbf{j}^{\mathrm{p}} = \mathbf{j}^{\mathrm{p}}_{\Psi}$ now, so the photon part of the wavefunction is unconstrained, hence the minimum of $\langle \Psi, H_{\mathrm{rad}} \Psi \rangle$ is zero.

Theorem (Zero coupling limit)

For any $(\rho, \mathbf{j}^{\mathrm{p}})$ N-representable such that $(\rho, \mathbf{j}^{\mathrm{p}} - e\mathbf{j}^{\mathrm{ph}})$ is also N-representable, the limit

$$F_{\mathrm{VR}}(\rho, \mathbf{j}^{\mathrm{p}}) = \lim_{e,\Lambda \to 0} F^{e,\Lambda}(\rho, \mathbf{j}^{\mathrm{p}} - e\mathbf{j}^{\mathrm{ph}})$$

holds true.

Quantum-electrodynamical DFT VII.

- **Existence of an optimizer?**
- The relevant space for the photon wavefunctions of finite kinetic energy is the **fractional order Sobolev space** $H^{1/2}$. (**Hopium:** There is Rellich–Kondrashov compact embedding into L^2 for this space.)
- **Representability?** Of $(\rho, \mathbf{j}^{\mathrm{c}})$ or $(\rho, \mathbf{j}^{\mathrm{p}}, \mathbf{j}^{\mathrm{ph}})$? We can prove it if $\boldsymbol{\omega}$ is divergence-free, $\rho\boldsymbol{\omega} = \mathbf{j}^{\mathrm{ph}}$.
- Introducing the **coupling function** (cf. Lieb–Loss)

$$h_{j,\nu}(x) = \int_{\mathbb{R}^3} \mathbb{1}_\Lambda(k) \frac{\varepsilon_j^\nu(k)}{\sqrt{|k|}} e^{-ik\cdot x}\,\mathrm{d}k$$

the vector potential operator my be written in position representation as

$$\mathfrak{A}^j(x) = \sum_{\nu=1,2} \int_{\mathbb{R}^3} \left(h_{j,\nu}(x-x')a^\dagger(x',\nu) + \overline{h_{j,\nu}(x-x')}a(x',\nu')\right)\mathrm{d}x'$$

$\Longrightarrow$ A kind of convolution operator. **The UV cutoff discards information.**

- Another issue is that $\mathbf{j}^{\mathrm{ph}}$ is a many-photon quantity.

QEDFT: Dressed density

- **Consider instead the "dressed" density?** For wavefunction $\Psi \in L^2_a(\mathbb{R}^{3N}, \mathcal{F}_{\mathrm{ph}})$ let
$$\boldsymbol{\rho}_\Psi(x_1) = \int_{\mathbb{R}^{3(N-1)}} \Psi(X) \otimes \overline{\Psi(X)} \,\mathrm{d}x_2 \dots \mathrm{d}x_N \in \mathfrak{S}_1(\mathcal{F}_{\mathrm{ph}})$$
which has $\boldsymbol{\rho}_\Psi(x_1) = \boldsymbol{\rho}_\Psi(x_1)^\dagger \geq 0$ as operators. Moreover, $\rho_\Psi = \mathrm{Tr}_{\mathcal{F}_{\mathrm{ph}}} \boldsymbol{\rho}_\Psi$.
- Then $\mathbf{j}^{\mathrm{ph}}_\Psi(x) = 2N \,\mathrm{Tr}_{\mathcal{F}_{\mathrm{ph}}} \mathfrak{A}(x)\boldsymbol{\rho}_\Psi(x)$
- Also $\langle \Psi, H_{\mathrm{rad}}\Psi\rangle = \mathrm{Tr}_{\mathcal{F}_{\mathrm{ph}}} H_{\mathrm{rad}}\boldsymbol{\rho}_\Psi(x)$
- So with dressed-density functional
$$F^{e,\Lambda}(\boldsymbol{\rho}, \mathbf{j}^{\mathrm{p}}) = \inf_{\substack{\Psi \in \mathfrak{H}^N_a \otimes \mathcal{F}_{\mathrm{ph}}, \|\Psi\|=1 \\ \boldsymbol{\rho}_\Psi = \boldsymbol{\rho} \\ \mathbf{j}^{\mathrm{p}}_\Psi = \mathbf{j}^{\mathrm{p}}}} \langle \Psi, H^{0,0}\Psi\rangle$$
we have

$$E_0(v, \mathbf{A}) = \inf_{\boldsymbol{\rho}, \mathbf{j}^{\mathrm{p}}} \left[F^{e,\Lambda}(\boldsymbol{\rho}, \mathbf{j}^{\mathrm{p}}) + \langle v + \tfrac{1}{2}|\mathbf{A}|^2, \rho\rangle + \langle \mathbf{A}, \mathbf{j}^{\mathrm{p}}\rangle - 2Ne \int_{\mathbb{R}^3} \mathbf{A}(x) \cdot \mathrm{Tr}_{\mathcal{F}_{\mathrm{ph}}} \mathfrak{A}(x)\boldsymbol{\rho}(x) \,\mathrm{d}x \right]$$

QEDFT: Truncation of the photon Fock space

- We first replace the one-photon Hilbert space $\mathfrak{H}_{\mathrm{ph}} = L^2(\mathbb{R}^3, \mathbb{C}^2)$ by

$$\mathfrak{H}_{\mathrm{ph}}^{\Lambda,\ell} = L^2(L_{\ell,\Lambda}, \mathbb{C}^2) \simeq \ell^2(L_{\ell,\Lambda}, \mathbb{C}^2) \simeq \mathbb{C}^K,$$

  where $L_{\ell,\Lambda} = (\ell\mathbb{Z}^3 \cap [-\Lambda, \Lambda]^3) \setminus \{0\}$ is equipped with the counting measure and $K \in \mathbb{N}$.
- Next, we truncate the maximum number of photons to $M_{\mathrm{ph}} \in \mathbb{N}$, and hence introduce the truncated photon Fock space

$$\mathcal{F}_{\mathrm{ph}}^{\leq N_{\mathrm{ph}},K} = \mathcal{F}_s^{\leq N_{\mathrm{ph}}}(\mathfrak{H}_{\mathrm{ph}}^K) \simeq \mathbb{C}^M$$

  for some $M \in \mathbb{N}$.
- The free field Hamiltonian becomes

$$H_{\mathrm{rad}} = \sum_{\nu=1,2} \sum_{k \in L} |k| \, a^\dagger(k,\nu) a(k,\nu)$$

- The vector potential operator becomes

$$\mathfrak{A}^j(x) = \sum_{\nu=1,2} \sum_{k \in L} \frac{\varepsilon_\nu^j(k)}{\sqrt{|k|}} \left[e^{-ik\cdot x} a^\dagger(k,\nu) + e^{ik\cdot x} a(k,\nu) \right]$$

Dressed DFT

Dressed DFT

- The density is a positive, Hermitian $\mathbb{C}^{m\times m}$-valued mapping $\boldsymbol{\rho} : \mathbb{R}^3 \to \mathbb{C}^{m\times m}$. This can come from either
  1. The internal degrees of freedom of the electrons (e.g. spin), or
  2. The photon field (or both).
- Such matrix-valued maps are in general **cannot be diagonalized globally**, due to topological obstructions and/or conical sections.
- The scalar Hoffmann–Ostenhof bound holds:

$$\int_{\mathbb{R}^3} |\boldsymbol{\nabla}\sqrt{\rho_\gamma}|^2 \leq \mathrm{Tr}(-\boldsymbol{\Delta}\gamma)$$

- Conjecture: The **generalized Hoffmann–Ostenhof bound** holds:

$$\int_{\mathbb{R}^3} |\boldsymbol{\nabla}\sqrt{\boldsymbol{\rho}_\gamma}|^2 \leq \mathrm{Tr}(-\boldsymbol{\Delta}\gamma)$$

Thank you for your attention!

Questions?