Quantum-Electrodynamical Density-Functional Theory

for the Dicke Hamiltonian

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Acknowledgements

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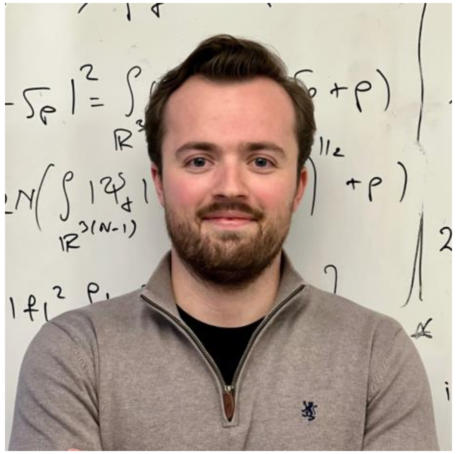


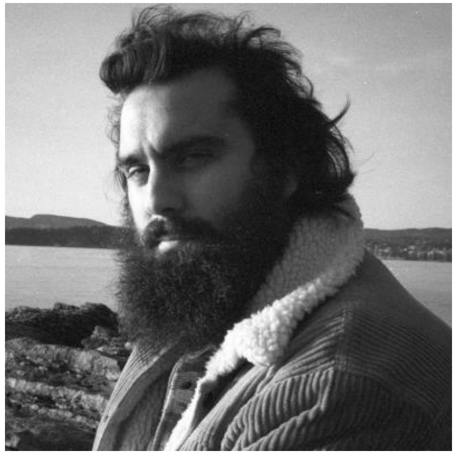


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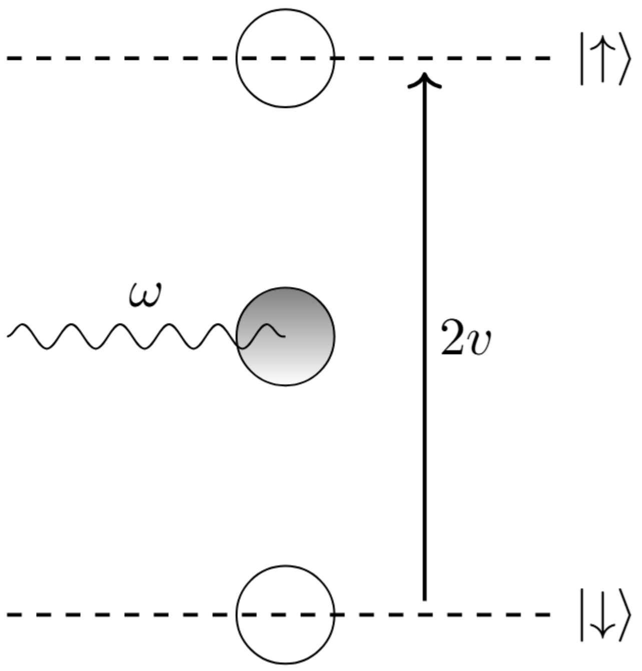
Motivation

- Importance of light-matter interactions $\Longrightarrow$
  QED = how charged particles interact through coupling to a quantum field
- Simple model (that can be extended)
- Study ground-state effects of coupling photons to electronic systems
- Studying an (almost) explicit form of a DFT functional: QEDFT

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



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

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

Examples

Let $N = 2$, then

$$\boldsymbol{\sigma}_z = \left(\begin{pmatrix} 1 & & & \\ & 1 & & \\ & & -1 & \\ & & & -1 \end{pmatrix}, \begin{pmatrix} 1 & & & \\ & -1 & & \\ & & 1 & \\ & & & -1 \end{pmatrix} \right)^\top$$

has always diagonal form and

$$\boldsymbol{\sigma}_x = \left(\begin{pmatrix} 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \\ 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \end{pmatrix}, \begin{pmatrix} 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{pmatrix} \right)^\top$$

Dicke Hamiltonian

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

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

Full Hamiltonian

$$\mathbf{H}(\mathbf{v}, \mathbf{j}) = \mathbf{H}_0 + \mathbf{v} \cdot \boldsymbol{\sigma}_z + \mathbf{j} \cdot \mathbf{x} \qquad \mathbf{v} \in \mathbb{R}^N, \qquad \mathbf{j} \in \mathbb{R}^M$$

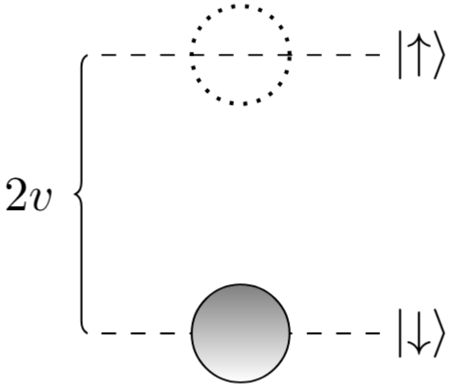
Hilbert space: $\mathcal{H} = \mathcal{H}_{\mathrm{ph}} \otimes \mathcal{H}_{\mathrm{f}}$

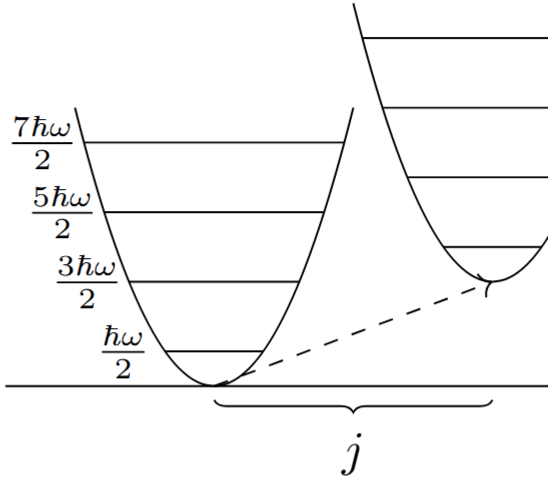
$\mathcal{H}_{\mathrm{ph}} = \bigotimes^M L^2(\mathbb{R})$ and $\mathcal{H}_{\mathrm{f}} = \bigotimes^N \mathbb{C}^2 \simeq \mathbb{C}^{2^N}$

Ground-state energy

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

$$Q_0 := Q(\mathbf{H}_0)$$





Internal “density” variables

Definition (Magnetization vector and photon coordinate)

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

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

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

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*

$$\sum_{n=1}^{N} (\boldsymbol{\sigma}_z^n)_{\boldsymbol{\alpha}\boldsymbol{\alpha}} (v_n^{(1)} - v_n^{(2)}) = E(\mathbf{v}^{(1)}, \mathbf{j}) - E(\mathbf{v}^{(2)}, \mathbf{j}), \tag{5}$$

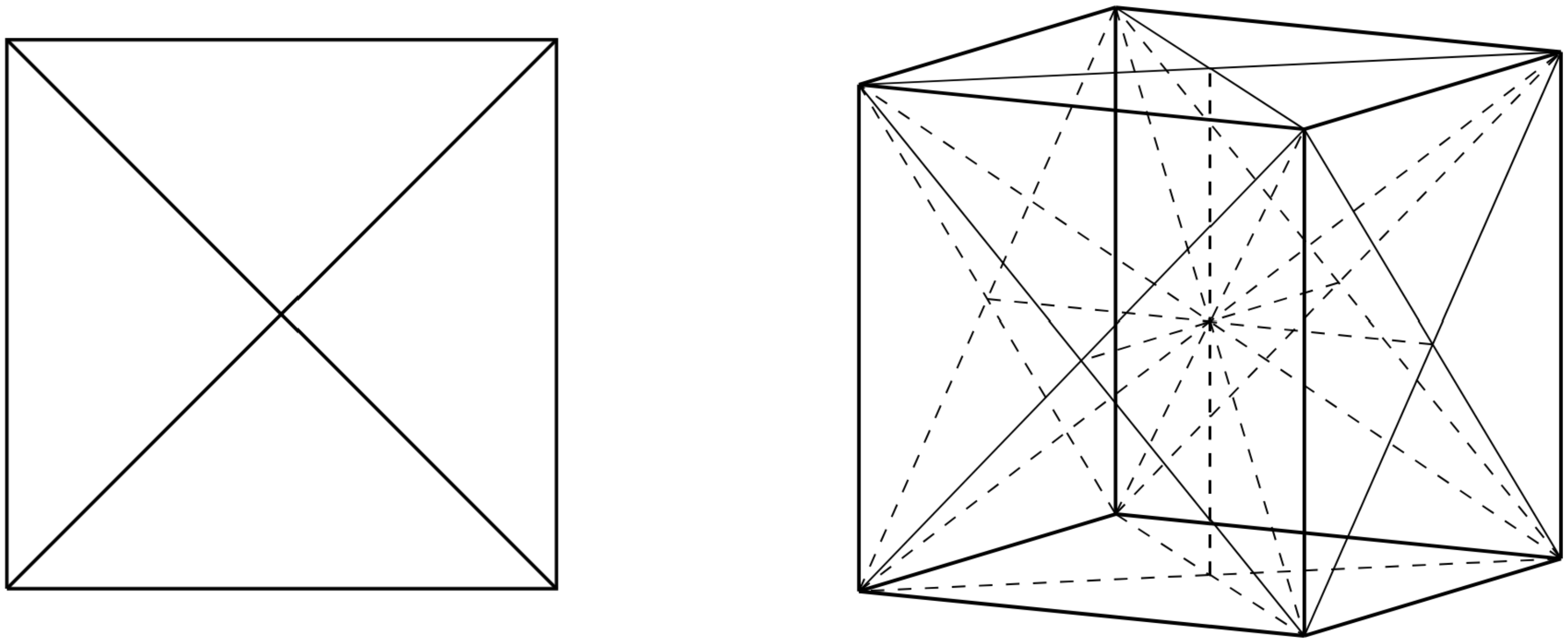
where $I^{(i)}$ *denotes the set of spinor indices* $\boldsymbol{\alpha}$ *for which* $(\psi^{(i)})^{\boldsymbol{\alpha}} \not\equiv 0$.

Regular case: Example

$N = 1$. $\mathcal{R}_1 = (-1, 1)$

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

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



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 (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

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

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

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*

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

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*

$$\mathbf{H}(\mathbf{v}, \mathbf{j})\psi = E\psi \tag{11}$$

and the second-order condition

$$\langle \boldsymbol{\chi}, \mathbf{H}(\mathbf{v}, \mathbf{j})\boldsymbol{\chi} \rangle \geq E\|\boldsymbol{\chi}\|^2, \tag{12}$$

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

$$F_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi}) = \langle \psi, \mathbf{H}_0 \psi \rangle = E - \mathbf{v} \cdot \boldsymbol{\sigma} - \mathbf{j} \cdot \boldsymbol{\xi}. \tag{13}$$

The second-order information (12) 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* (11) *and* (12) *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”.

$M = N = 1$

Corollary 4.8. *Consider a regular density pair* $(\sigma, \xi) \in (-1, 1) \times \mathbb{R}$. *Then the following holds:*

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

2. *(equivalence of functionals)* $F_{\mathrm{LL}}(\sigma, \xi) = F_{\mathrm{L}}(\sigma, \xi)$.

3. *(differentiability) The* F_{LL} *is differentiable at* (σ, ξ) *and* $(v, j) = -\nabla F_{\mathrm{LL}}(\sigma, \xi)$ *are its representing external potentials.*

Summary

- Study of an (almost) explicit form of a DFT
- Sharper results on Hohenberg–Kohn and v-rep
- More direct properties of the functional (to be used in future work)

Availabe on arXiv

QUANTUM-ELECTRODYNAMICAL DENSITY-FUNCTIONAL THEORY FOR THE DICKE HAMILTONIAN

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ABSTRACT. A detailed analysis of density-functional theory for quantum-electrodynamical model systems is provided. In particular, the quantum Rabi model, the Dicke model, and a generalization of the latter to multiple modes are considered. We prove a Hohenberg–Kohn theorem that manifests the magnetization and displacement as internal variables, along with several representability results. The constrained-search functionals for pure states and ensembles are introduced and analyzed. We find the optimizers for the pure-state constrained-search functional to be low-lying eigenstates of the Hamiltonian and, based on the properties of the optimizers, we formulate an adiabatic-connection formula. In the reduced case of the Rabi model we can even show differentiability of the universal density functional, which amounts to unique pure-state v-representability.

Thank you for your attention!