Quantum-Electrodynamical Density-Functional Theory

Exemplified by the Multimode Dicke Model

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Acknowledgements

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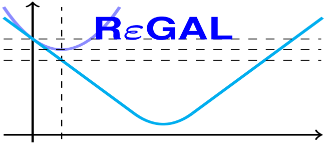

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Quantum-Electrodynamical Density-Functional Theory Exemplified by the Quantum Rabi Model

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The key features of density-functional theory (DFT) within a minimal implementation of quantum electrodynamics are demonstrated, thus allowing to study elementary properties of quantum-electrodynamical density-functional theory (QEDFT). We primarily employ the quantum Rabi model, that describes a two-level system coupled to a single photon mode, and also discuss the Dicke model, where multiple two-level systems couple to the same photon mode. In these settings, the density variables of the system are the polarization and the displacement of the photon field. We give analytical expressions for the constrained-search functional and the exchange-correlation potential and compare to established results from QEDFT. We further derive a form for the adiabatic connection that is almost explicit in the density variables, up to only a non-explicit correlation term that gets bounded both analytically and numerically. This allows several key features of DFT to be studied without approximations.

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I. INTRODUCTION

A. Prelude and Overview

The study of light-matter interactions forms the basis for understanding a wide range of phenomena and those effects are instrumental for measuring and manipulating matter in experiments. At the fundamental level, charged particles interact among each other through their coupling to the photon field, a process that is described by quantum electrodynamics (QED) [1–6]. While the quantization of the electromagnetic field is often considered to only be relevant for high-energy physics, QED effects, such as spontaneous emission or the Purcell effect, also occur in the low-energy (non-relativistic)

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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.

1. Introduction

Quantum electrodynamics (QED) is the fully quantized theory of matter and light [Ryd96; GR13]. It describes the interaction between charged particles through their coupling to the electromagnetic field. Apart from high-energy physics, particularly in the domain of equilibrium condensed-matter physics, non-relativistic QED

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State Space & Lifted Pauli Matrices

N two-level systems and M QHOs:

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

$$\sigma_a^j = \mathbb{1} \otimes \cdots \otimes \mathbb{1} \otimes \underbrace{\sigma_a}_{j\text{th}} \otimes \mathbb{1} \otimes \cdots \otimes \mathbb{1}, \quad a \in \{x, y, z\}$$

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \qquad \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \qquad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

$$\boldsymbol{\sigma}_a = \begin{pmatrix} \sigma_a^1 \\ \sigma_a^2 \\ \vdots \\ \sigma_a^N \end{pmatrix} \in \left(\mathbb{C}^{2^N \times 2^N}\right)^N$$

Example: $N = 2$

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

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

$$\langle \boldsymbol{\psi}, \boldsymbol{\sigma}_z^1 \boldsymbol{\psi} \rangle = \|\psi_{++}\|^2_{L^2(\mathbb{R}^M)} + \|\psi_{+-}\|^2_{L^2(\mathbb{R}^M)} - \|\psi_{-+}\|^2_{L^2(\mathbb{R}^M)} - \|\psi_{--}\|^2_{L^2(\mathbb{R}^M)}$$

The Multimode Dicke Hamiltonian

Internal Hamiltonian

NB! Math units, e.g., $\frac{\omega}{2} = 1$

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

$$\Lambda \in \mathbb{R}^{M \times N} \qquad \mathbf{t} \in \mathbb{R}^N \quad (\mathbf{t} \neq 0)$$

"Potentials":

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

Full Hamiltonian

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

"Densities":

$$\boldsymbol{\sigma} = \langle \psi,\, \boldsymbol{\sigma}_z \psi \rangle \in [-1, 1]^N \qquad \boldsymbol{\xi} = \langle \psi,\, \mathbf{x}\psi \rangle \in \mathbb{R}^M$$

A Density-Functional Theory

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The Ground-State Problem

$$Q_0 := Q(\mathbf{H}_0) \quad \longleftarrow \quad \text{form domain of QHO}$$

$$E(\mathbf{v},\mathbf{j}) = \inf_{\substack{\boldsymbol{\psi}\in Q_0 \\ \|\boldsymbol{\psi}\|^2=1}} \langle \boldsymbol{\psi},\, \mathbf{H}(\mathbf{v},\mathbf{j})\boldsymbol{\psi}\rangle$$

Theorem (N-representability)

For every $(\boldsymbol{\sigma},\boldsymbol{\xi}) \in [-1,1]^N \times \mathbb{R}^M$ there exists a $\psi \in Q_0$ such that

$$\|\boldsymbol{\psi}\| = 1, \quad \langle \boldsymbol{\psi},\, \boldsymbol{\sigma}_z\boldsymbol{\psi}\rangle = \boldsymbol{\sigma}, \quad \textit{and} \quad \langle \boldsymbol{\psi},\, \mathbf{x}\boldsymbol{\psi}\rangle = \boldsymbol{\xi}.$$

Regular Density

Definition (Regular Polarisation [1])

$\boldsymbol{\sigma} \in [-1,1]^N$ is called *regular* if for every $\chi \in \mathbb{R}_+^{2^N}$ such that

$$\|\chi\| = 1 \quad \text{and} \quad \langle \chi, \boldsymbol{\sigma}_z \chi \rangle = \boldsymbol{\sigma}$$

one has $\{\chi, \sigma_z^1 \chi, \dots, \sigma_z^N \chi\}$ as a set of linear independent vectors.

The set of all regular $\boldsymbol{\sigma}$ is denoted $\mathcal{R}_N$.

Any $\boldsymbol{\sigma} \in [-1,1]^N \setminus \mathcal{R}_N$ is *not regular*.

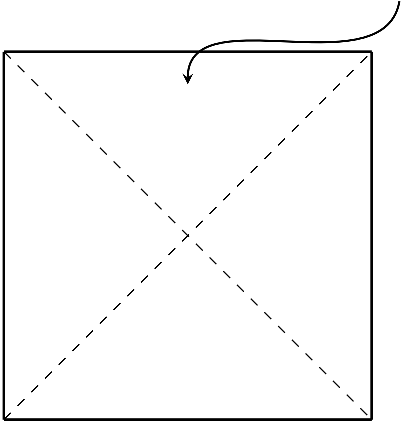
Example: Regular Density

If $N = 1$,

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

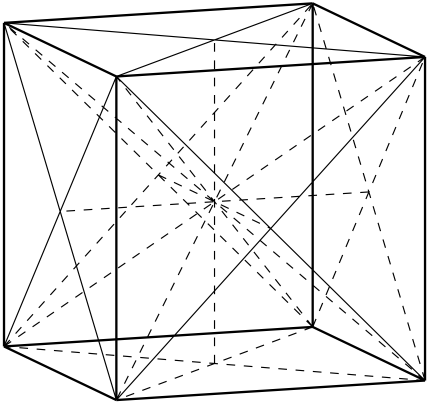
If $N = 2$,

$$\mathcal{R}_2 \subset (-1, 1)^2$$



Example: Regular Density $N = 3$

$$\mathcal{R}_3 \subset (-1,1)^3$$



$\mathcal{R}_N$: union of disjoint open convex polytopes

$[-1,1]^N \setminus \mathcal{R}_N$: union of finite number of hyperplanes intersected with $[-1,1]^N$

A Hohenberg–Kohn Theorem

Theorem

Any density pair $(\boldsymbol{\sigma}, \boldsymbol{\xi}) \in \mathcal{R}_N \times \mathbb{R}^M$ *of a ground state uniquely determines an external pair* $(\mathbf{v}, \mathbf{j}) \in \mathbb{R}^N \times \mathbb{R}^M$*. That is, the mapping*

$$\mathbb{R}^N \times \mathbb{R}^M \ni (\mathbf{v}, \mathbf{j}) \longmapsto (\boldsymbol{\sigma}, \boldsymbol{\xi}) \in \mathcal{R}_N \times \mathbb{R}^M$$

is an injection.

Internal variables

$$(\boldsymbol{\sigma}, \boldsymbol{\xi}) \in [-1, 1]^N \times \mathbb{R}^M$$

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The Levy–Lieb Functional

Constraint manifold

$$\mathcal{M}_{\boldsymbol{\sigma},\boldsymbol{\xi}} = \{\psi \in Q_0 : \|\psi\| = 1,\ \langle\psi,\ \boldsymbol{\sigma}_z\psi\rangle = \boldsymbol{\sigma},\ \langle\psi,\ \mathbf{x}\psi\rangle = \boldsymbol{\xi}\}$$

$F_{\mathrm{LL}} : [-1,1]^N \times \mathbb{R}^M \to \mathbb{R}$,

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

$$E(\mathbf{v},\mathbf{j}) = \inf_{(\boldsymbol{\sigma},\boldsymbol{\xi})\in[-1,1]^N\times\mathbb{R}^M} \{F_{\mathrm{LL}}(\boldsymbol{\sigma},\boldsymbol{\xi}) + \mathbf{v}\cdot\boldsymbol{\sigma} + \mathbf{j}\cdot\boldsymbol{\xi}\}$$

Properties of F_{LL}

For every $(\boldsymbol{\sigma}, \boldsymbol{\xi}) \in [-1,1]^N \times \mathbb{R}^M$

- *Symmetric*

$$F_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi}) = F_{\mathrm{LL}}(-\boldsymbol{\sigma}, -\boldsymbol{\xi})$$

- *Displacement rule*

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

- There exists a *real-valued optimiser* of $F_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi})$
- *Virial relation*: for any optimiser ψ of $F_{\mathrm{LL}}(\boldsymbol{\sigma}, 0)$,

$$\|\boldsymbol{\nabla}\psi\|^2 - \|\mathbf{x}\psi\|^2 = \frac{1}{2}\langle \psi,\, \mathbf{x} \cdot \Lambda \boldsymbol{\sigma}_z \psi \rangle$$

Optimiser of F_{LL}

Theorem (Optimisers are low-lying eigenstates)

Let $(\boldsymbol{\sigma}, \boldsymbol{\xi}) \in \mathcal{R}_N \times \mathbb{R}^M$. *Suppose that* $\psi \in \mathcal{M}_{\boldsymbol{\sigma},\boldsymbol{\xi}}$ *is an optimiser of* $F_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi})$ *with Lagrange multipliers* $E \in \mathbb{R}$, $\mathbf{v} \in \mathbb{R}^N$, *and* $\mathbf{j} \in \mathbb{R}^M$ *such that* ψ *satisfies*

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

and for all χ *in the tangent space of* $\mathcal{M}_{\boldsymbol{\sigma},\boldsymbol{\xi}}$ *at* ψ,

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

Then ψ *is at most the* $(N + M)$*th excited eigenstate of* $\mathbf{H}(\mathbf{v}, \mathbf{j})$, *and*

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

Conclusions

- Very explicit form of DFT
- Regular densities
- Characterisation of optimisers

Open questions ($N \geq 2$):

- v-representability
- Differentiability of F_{LL}
- Does $F_{\mathrm{LL}} = F_{\mathrm{L}}$?

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Thank you for your attention!
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Appendix

Motivation

- Light-matter interactions
- Ground-state effects of photon-electron coupling
- Explicit from (almost) of a DFT functional
- Simple model

The Quantum Rabi Model

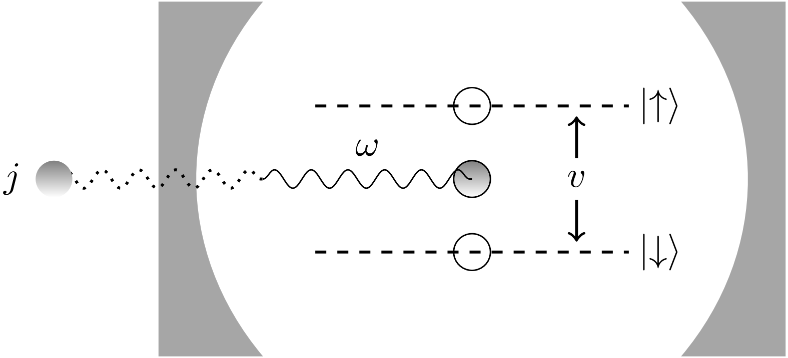
$$\widehat{H}_0 = \underbrace{\frac{1}{2}\widehat{p}^2 + \frac{\omega^2}{2}\widehat{x}^2}_{\text{QHO}} \underbrace{-t\widehat{\sigma}_x}_{\text{TLS kin.}} + \underbrace{g\widehat{\sigma}_z\widehat{x}}_{\text{TLS-QHO coupling}}$$

$$\psi \in \mathbb{C}^2 \otimes L^2(\mathbb{R},\mathbb{C}) \simeq L^2(\mathbb{R},\mathbb{C}^2)$$

$$[-1,1] \ni \sigma := \langle \psi,\, \widehat{\sigma}_z \psi \rangle$$

$$\widehat{H}(v,j) = \widehat{H}_0 + v\widehat{\sigma}_z + j\widehat{x}$$

$$\mathbb{R} \ni \xi := \langle \psi,\, \widehat{x}\psi \rangle$$



Coupling Strength Functional

Coupling strength parameter: $s \in \mathbb{R}$

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

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

Q_0 unchanged

A Trial State

Lemma

Let $(\boldsymbol{\sigma}, \boldsymbol{\xi}) \in [-1,1]^N \times \mathbb{R}^M$ *and* $\Lambda = 0$*, then the state*

$$\psi(x) = \frac{1}{\pi^{M/4}} e^{-\frac{1}{2}|\mathbf{x}-\boldsymbol{\xi}|^2} \mathbf{c}, \qquad \mathbf{c} = \bigotimes_{n=1}^{N} \begin{pmatrix} \sqrt{\frac{1+\sigma_n}{\sqrt{2}}} \\ \sqrt{\frac{1-\sigma_n}{\sqrt{2}}} \end{pmatrix}$$

is an optimiser of $F^0_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi})$*, and*

$$F^0_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi}) = M + |\boldsymbol{\xi}|^2 - \sum_{n}^{N} t_n \sqrt{1 - \sigma_n^2}.$$

The Superdifferential of F_{LL}

$$\mathbb{R}^{M\times N} \ni \Lambda \longmapsto F_{\mathrm{LL}}^{\Lambda}(\boldsymbol{\sigma},\boldsymbol{\xi}) \quad \text{and} \quad \mathbb{R} \ni s \longmapsto F_{\mathrm{LL}}^{s\Lambda}(\boldsymbol{\sigma},\boldsymbol{\xi}) \quad \text{convex}$$

Lemma

For every (fixed) $(\boldsymbol{\sigma},\boldsymbol{\xi}) \in [-1,1]^N \times \mathbb{R}^M$ *and* $s \in \mathbb{R}$*, then*

$$\overline{\partial}_s \big[F_{\mathrm{LL}}^{s\Lambda}(\boldsymbol{\sigma},\boldsymbol{\xi})\big]\,(s) \supset \big\{\langle\psi_s,\, \mathbf{x}\cdot\Lambda\boldsymbol{\sigma}\psi_s\rangle : \psi_s \in Q_0 \text{ with } F_{\mathrm{LL}}^{s\Lambda} = \langle\psi_s,\, \mathbf{H}_0^{s\Lambda}\psi_s\rangle\big\}$$

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The Adiabatic Connection

Theorem

The functional $F^{\Lambda}_{\mathrm{LL}} : [-1,1]^N \times \mathbb{R}^M \to \mathbb{R}$ *satisfies*

$$F^{\Lambda}_{\mathrm{LL}}(\boldsymbol{\sigma}, \boldsymbol{\xi}) = M + |\boldsymbol{\xi}|^2 - \sum_n^N t_n \sqrt{1 - \sigma_n^2} + \boldsymbol{\xi} \cdot \Lambda \boldsymbol{\sigma} + G^{\Lambda}(\boldsymbol{\sigma}).$$

Here

$$G^{\Lambda}(\boldsymbol{\sigma}) := \frac{1}{2}|\Lambda \boldsymbol{\sigma}|^2 - \int_0^1 \left(\frac{1}{2} \|\Lambda \boldsymbol{\sigma}_z \psi_s\|^2 + \langle \mathbf{t} \cdot \boldsymbol{\sigma}_x \psi_s, \boldsymbol{\nabla} \cdot \Lambda(\boldsymbol{\sigma}_z - \boldsymbol{\sigma}) \psi_s \rangle \right) \mathrm{d}s \, ,$$

and $\psi_s \in \mathcal{M}_{\boldsymbol{\sigma},0}$ *is a real-valued optimiser of* $F^{s\Lambda}_{\mathrm{LL}}(\boldsymbol{\sigma}, 0)$.

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