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Density-Functional Theory Exemplified

By Application to a Quantum Electrodynamical Model

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OSLO METROPOLITAN UNIVERSITY
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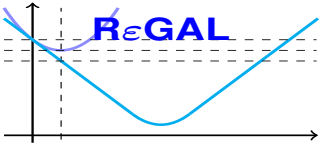
Acknowledgements

With A. Csirik,[1] M. Penz,[1,2,3,4,5] V. Falmår,[1] M. Lotfigolian,[1] M. Ruggenthaler,[4,5] and A. Laestadius.[1,6]



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Article

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

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Vebjørn H. Bakkestuen, Vegard Falmår, Maryam Lotfigolian, Markus Penz, Michael Ruggenthaler, and Andre Laestadius*

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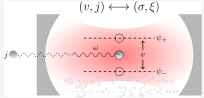
ABSTRACT: The key features of density-functional theory (DFT) within a minimalistic implementation of quantum electrodynamics are demonstrated, thus allowing to study elementary properties of quantum-electrodynamical density-functional theory (QEDFT). We primarily employ the quantum Rabi model that describes 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 them 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 nonexplicit correlation term that gets bounded both analytically and numerically. This allows several key features of DFT to be studied without approximations.



1. INTRODUCTION

1.1. Prelude and Overview. The study of light−matter interactions forms the basis for understanding a wide range of phenomena whose 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).[1−4] 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 (nonrelativistic) regime of charged particles. In recent years, many experimental and theoretical works have shown that in optical environments, such as Fabry−Pérot cavities, changes in the quantized light field can modify chemical and material properties even at equilibrium.[5−9] It has therefore become increasingly relevant to extend well-established first-principles methods, such as density-functional theory (DFT) and coupled-cluster theory, to encompass QED.[10−13]

Due to its computational simplicity, DFT is the method predominantly used when studying quantum systems with large numbers of particles. In DFT, the N-body wave function—with its intractable dimensionality—is replaced by the one-body particle density. This dimensional reduction is precisely why DFT calculations provide effective approximations and thus have become an indispensable tool across many fields, such as chemistry, materials science and solid-state physics.[14−17] Since the seminal papers of Hohenberg and Kohn,[18] Kohn and Sham,[19] and Lieb,[20] significant efforts have been devoted both to the numerical and mathematical developments of DFT. Besides, motivated by its extremely elegant formulation in terms of convex analysis, a perspective toward DFT has emerged that makes it more than just an approximation method. The concave form of the ground-state energy in terms of the external potential naturally yields the universal functional as its Legendre−Fenchel transform. Then the Hohenberg−Kohn theorem is the statement that the subdifferential of the universal functional just contains a single element (see also Section 1.4) and v-representability connects closely to differentiability of this functional. The whole of DFT thus follows as a convex treatment of many-body quantum mechanics in the ground state. In this sense, DFT can be referred to more as a discovery than an invention.[21] The DFT

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Density-Functional Theory for the Dicke Hamiltonian

Vebjørn H. Bakkestuen [1] · Mihály A. Csirik [1,2] · Andre Laestadius [1,2] · Markus Penz [1,3]

Received: 26 September 2024 / Accepted: 29 March 2025

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.

Keywords Non-relativistic quantum electrodynamics · Rabi model · Dicke model · Density-functional theory · Hohenberg–Kohn theorem · Representability · Mathematical physics

1 Introduction

Quantum electrodynamics (QED) is the fully quantized theory of matter and light [17, 47]. 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 in the shape of the Pauli–Fierz Hamiltonian [52] is considered sufficient to describe interesting effects, such as the modification of chemical and material properties [12, 15, 45]. In order to explain those, and due to the high number of involved particles and ensuing complexity of the problem, well-established first-principle approximation methods, such as density-functional theory (DFT), were adapted for

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Motivation

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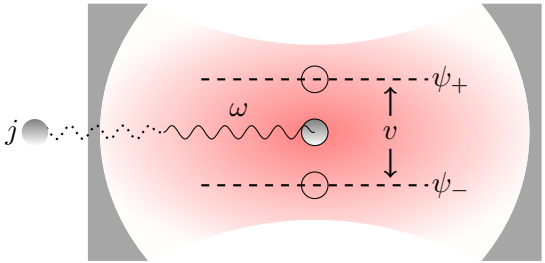
The Quantum Rabi Model

$$\widehat{H}_0^\lambda = \frac{1}{2}\widehat{p}^2 + \frac{\omega^2}{2}\widehat{q}^2 + \lambda g \widehat{\sigma}_z \widehat{q} - t\widehat{\sigma}_x$$



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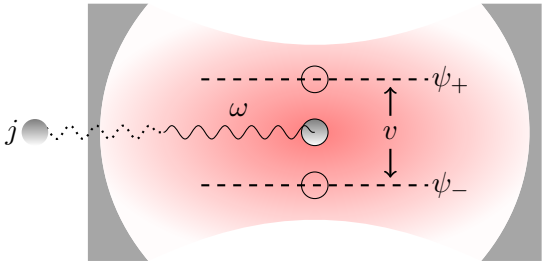

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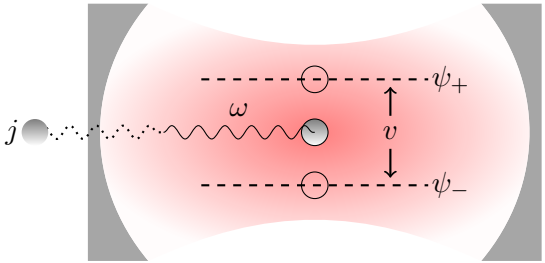

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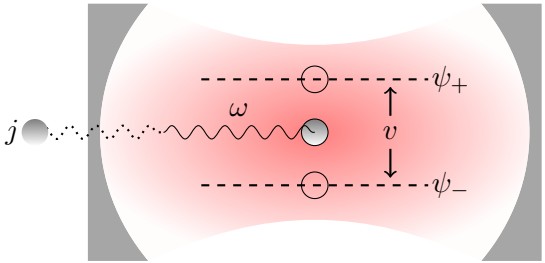

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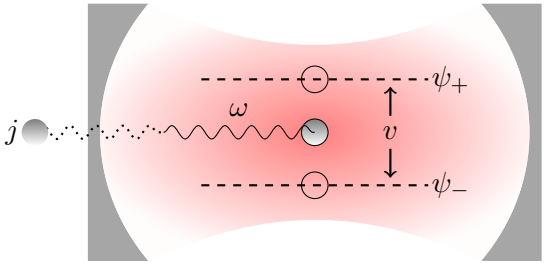

$$\sigma := \langle\psi|\widehat{\sigma}_z|\psi\rangle \in [-1,1]$$

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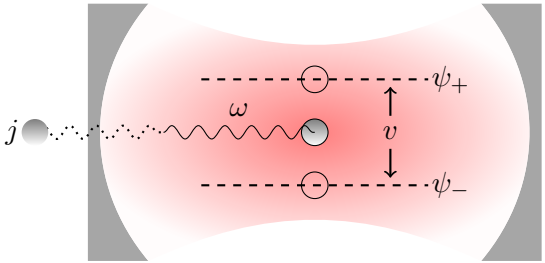

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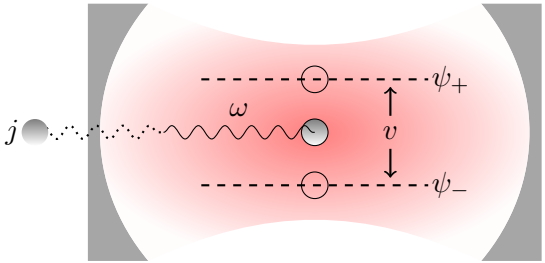

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"Density space": $X = [-1,1] \times \mathbb{R}$

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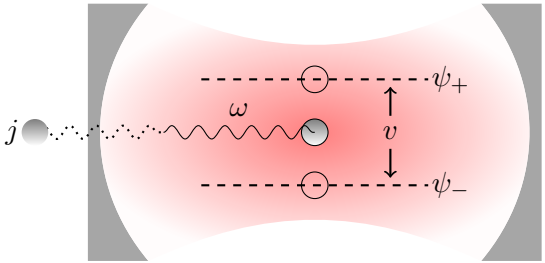

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$$\psi \longmapsto (\sigma,\xi) \in X$$

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Density-Functional Theory

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The Hohenberg–Kohn Theorem I

Lemma (Weak Hohenberg–Kohn theorem (HK1))

Suppose $\psi_1, \psi_2 \in Q_0$ are ground states of $\widehat{H}(v_1, j_1)$ and $\widehat{H}(v_2, j_2)$, respectively. If both $\psi_1, \psi_2 \mapsto (\sigma, \xi)$, then ψ_2 is a ground state of $\widehat{H}(v_1, j_1)$ and ψ_1 is a ground state of $\widehat{H}(v_2, j_2)$.

The Hohenberg–Kohn Theorem I

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Lemma (HK2)

If two Hamiltonians $\widehat{H}(v_1, j_1)$ *and* $\widehat{H}(v_2, j_2)$ *share any ground state then* $v_1 = v_2$ *and* $j_1 = j_2$*.*

The Hohenberg–Kohn Theorem II

Theorem (Standard Hohenberg–Kohn Theorem [3])

For an electronic system, the ground-state density $\rho(\mathbf{r})$ determines the potential $v(\mathbf{r})$ up to an arbitrary additive constant.

$$\rho(\mathbf{r}) \longmapsto v(\mathbf{r}) + \text{const.}$$

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Theorem (Hohenberg–Kohn Theorem for QRabi)

Any density pair $(\sigma, \xi) \in (-1, 1) \times \mathbb{R}$ uniquely determines an external pair $(v, j) \in \mathbb{R}^2$. That is, the Hohenberg–Kohn mapping

$$(-1, 1) \times \mathbb{R} \ni (\sigma, \xi) \longmapsto (v, j) \in \mathbb{R}^2$$

is a bijection.

The Levy–Lieb Functional

$$\mathcal{M}_{\sigma,\xi} := \{\psi \in Q_0 : \|\psi\| = 1,\ \psi \mapsto (\sigma, \xi)\}$$

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$$\begin{aligned} E^{\lambda}(v,j) &= \inf_{\psi \in Q_0} \langle \psi | \widehat{H}^{\lambda}(v,j) | \psi \rangle \\ &= \inf_{(\sigma,\xi)\in X} \left[\inf_{\psi \in \mathcal{M}_{\sigma,\xi}} \langle \psi | \widehat{H}_0^{\lambda} | \psi \rangle + v\sigma + j\xi \right] \end{aligned}$$

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Levy–Lieb functional $F_{\mathrm{LL}}^\lambda : X \to \overline{\mathbb{R}} = \mathbb{R} \cup \{+\infty\}$

$$F_{\mathrm{LL}}^\lambda(\sigma,\xi) = \inf_{\psi \in \mathcal{M}_{\sigma,\xi}} \langle \psi | \widehat{H}_0^\lambda | \psi \rangle$$

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The Universality of the Density-Functional

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- the universal functional F^λ is differentiable at (σ,ξ), and

$$(v,j) = -\nabla F^\lambda(\sigma,\xi)$$

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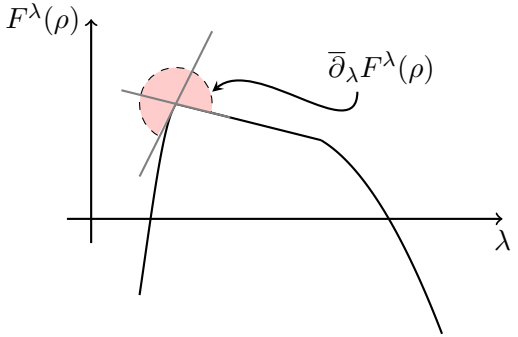
Mathematical Interlude

Given a concave functional $\lambda \mapsto F^\lambda$, the *superdifferential* of F^λ w.r.t. λ is

$$\overline{\partial}_\lambda F^\lambda(\rho) = \left\{ f \in \mathbb{R} \,|\, \forall \lambda' \in \mathbb{R} : F^{\lambda'}(\rho) \leq F^\lambda(\rho) + f \cdot \left(\lambda' - \lambda\right) \right\}$$



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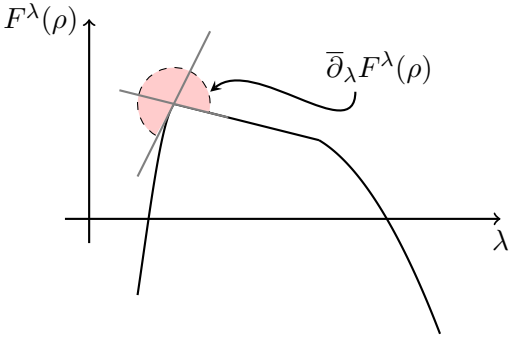
The generalised Newton–Leibniz formula

$$F^\lambda(\rho) = F^0(\rho) + \int_0^\lambda f^\mu(\rho)\, \mathrm{d}\mu$$

for any element $f^\lambda(\rho) \in \overline{\partial}_\lambda F^\lambda(\rho)$



The Adiabatic Connection Functional

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$$\begin{aligned} F^\lambda(\sigma,\xi) &= \overbrace{\frac{\omega}{2} - t\sqrt{1-\sigma^2} + \frac{\omega^2}{2}\xi^2}^{F^0(\sigma,\xi)} + \int_0^\lambda g\,\langle\psi^\mu|\widehat{\sigma}_z\widehat{q}|\psi^\mu\rangle\,\mathrm{d}\mu \\ &= \frac{\omega}{2} - t\sqrt{1-\sigma^2} + \frac{\omega^2}{2}\xi^2 + \underbrace{\lambda g\sigma\xi}_{D(\sigma,\xi)} - \frac{\lambda^2 g^2}{2\omega^2}(1-\sigma^2) - \frac{4t}{\omega^2}\int_0^\lambda\int_{\mathbb{R}} \varphi_+^{\mu\,\prime}\varphi_-^\mu\,\mathrm{d}q\,\mathrm{d}\mu \end{aligned}$$

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Standard DFT:

$$F^\lambda(\rho) = T(\rho) + \lambda E_{\mathrm{H}}(\rho) + \int_0^\lambda \Big(\mathrm{Tr}\Big[\widehat{W}\Gamma^\mu\Big] - E_{\mathrm{H}}(\rho)\Big)\,\mathrm{d}\mu$$

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Exchange Energy

Mathematical definition of exchange in DFT [4]

$$E_{\mathrm{x}}(\rho) := \lim_{\lambda \to 0^{+}} \frac{F^{\lambda}(\rho) - F^{0}(\rho)}{\lambda} - E_{\mathrm{H}}(\rho)$$

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$$E_{\mathrm{x}}(\sigma, \xi) = \lim_{\lambda \to 0^+} \frac{F^{\lambda}(\sigma, \xi) - F^{0}(\sigma, \xi)}{\lambda} - D(\sigma, \xi) = 0$$

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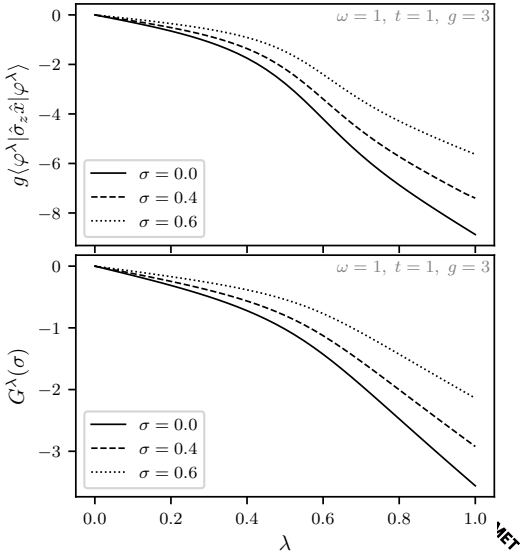

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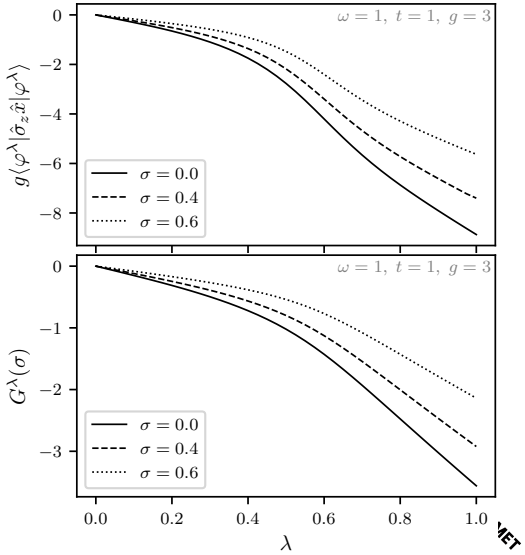
Conjecture [6]:

$$\overline{\partial}_{\lambda} F^{\lambda}(\rho) - E_{\mathrm{H}}(\rho) \ni f^{\lambda}(\rho) \quad \longleftarrow \text{ convex in } \lambda$$



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- v-representability

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

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Appendix

The Basic Operators

Quantum Harmonic Oscillator

$$\widehat{q} = \frac{1}{\sqrt{2\omega}}\left(\widehat{a}^\dagger + \widehat{a}\right) \quad \text{and} \quad \widehat{p} = i\sqrt{\frac{\omega}{2}}\left(\widehat{a}^\dagger - \widehat{a}\right)$$

$$\left[\tfrac{1}{2}\widehat{p}^2 + \tfrac{\omega^2}{2}\widehat{q}^2\right]|\psi^n\rangle = \omega\left[\tfrac{1}{2} + n\right]|\psi^n\rangle \quad \text{if } |\psi^n\rangle \text{ is the } n\text{th eigenstate}$$

Pauli-operators

$$\widehat{\sigma}_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \widehat{\sigma}_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \widehat{\sigma}_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.$$

N-Representability

For any $(\sigma, \xi) \in [-1, 1] \times \mathbb{R}$ there exists an admissable wavefunction ψ such that

$$\langle \psi | \widehat{\sigma}_z | \psi \rangle = \sigma \quad \text{and} \quad \langle \psi | \widehat{q} | \psi \rangle = \xi.$$

Proof by construction:

$$\psi(q) = \begin{pmatrix} c_+ \\ c_- \end{pmatrix} \sqrt[4]{\frac{\omega}{\pi}} e^{-\frac{\omega}{2}(q-\xi)^2}, \quad c_\pm = \sqrt{\frac{1 \pm \sigma}{2}}.$$

Properties of the QRabi Universal Functional

For every $(\sigma, \xi) \in [-1, 1] \times \mathbb{R}$ and any optimiser ψ of $F_{\mathrm{LL}}(\sigma, \xi)$, the following hold

1. $F(\sigma, \xi) = F(-\sigma, -\xi)$.
2. For any $\zeta \in \mathbb{R}$, $F^{\lambda}(\sigma, \xi + \zeta) = F^{\lambda}(\sigma, \xi) + \omega^2 \zeta(\xi + \frac{\zeta}{2}) + \lambda g \sigma \zeta$,
   for $\zeta = 0$, $F^{\lambda}(\sigma, \xi) = F^{\lambda}(\sigma, 0) + \lambda g \sigma \xi + \frac{\omega^2}{2}\xi^2$
3. ψ can always be chosen real and non-negative in both components ($\psi_{\pm}$).
4. Any optimiser ψ of $F_{\mathrm{LL}}(\sigma, 0)$ satisfies the virial relation

$$\int \left(\frac{1}{2} |\psi'|^2 - \frac{\omega^2}{2} q^2 |\psi|^2 \right) \mathrm{d}q = g \int q |\psi_+|^2 \, \mathrm{d}q \, .$$

5. For any optimiser

$$\int q |\psi_+|^2 \, \mathrm{d}q = \xi - \int q |\psi_-|^2 \, \mathrm{d}q = -\frac{2t}{\omega^2} \int \psi_+' \psi_- \, \mathrm{d}q - \frac{g(1 - \sigma^2)}{2\omega^2} + \frac{\xi(1 + \sigma)}{2}.$$

6. Any optimiser satisfies the bound $\int \psi_+'' \psi_- \, \mathrm{d}q \leq \frac{\omega^2}{8t}(1 - \sigma^2)$.

A Lieb–Oxford Bound for QRabi

The correlation energy satisfies the Lieb–Oxford-type bound

$$0 \geq \lambda E_{\mathrm{c}}^{\lambda}(\sigma) \geq -\frac{\lambda^2 g^2}{2\omega^2}(1-\sigma^2).$$

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Approximate Correlation for QRabi

$$I^{\lambda}(\sigma) := -\frac{4t}{\omega^2}\int_0^{\lambda}\int_{\mathbb{R}} {\varphi_+^{\mu}}'(q)\varphi_-^{\mu}(q)\,\mathrm{d}q\,\mathrm{d}\mu$$

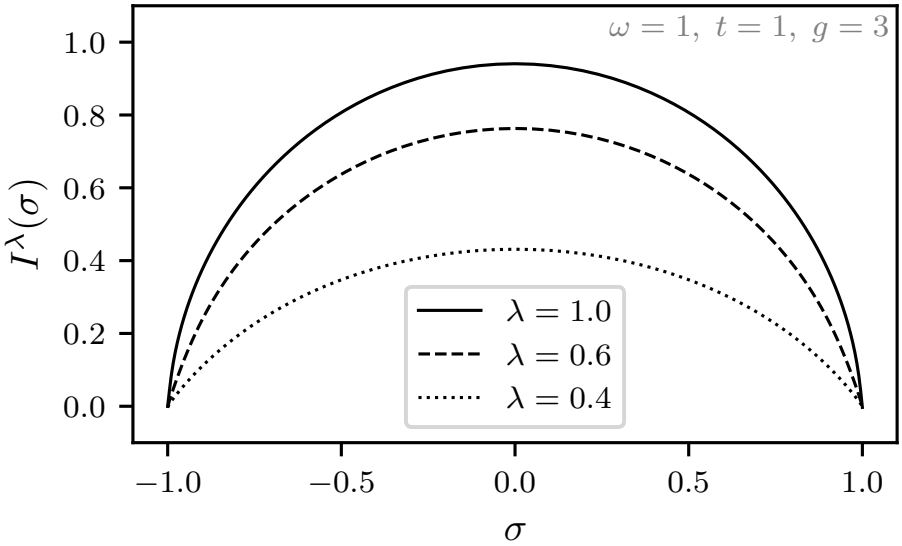
Conjecture

For $\sigma \in [-1,1]$ the non-explicit correlation functional is of the approximate form

$$I^{\lambda}(\sigma) \approx \omega\frac{b(\lambda,t)}{a(\lambda,t)}\Big[\sqrt{a(\lambda,t)^2-\sigma^2}-\sqrt{a(\lambda,t)^2-1}\Big].$$

Here a and b are functions of the parameters λ and t.

Non-Explicit Correlation for QRabi



The Coupling-Strength Density Functional

$$\widehat{T} = -\frac{1}{2}\sum_{j=1}^{N}\nabla_{\mathbf{r}_j}^2 \qquad \widehat{W} = \frac{1}{2}\sum_{j=1}^{N}\sum_{k\neq j}^{N}\frac{1}{|\mathbf{r}_j - \mathbf{r}_k|} \qquad \widehat{V} = \sum_{j=1}^{N} v(\mathbf{r}_j)$$

$$E^{\lambda}(v) = \inf_{\psi}\ \langle\psi|\widehat{T} + \lambda\widehat{W} + \widehat{V}|\psi\rangle \qquad F^{\lambda}(\rho) = \sup_{v}\Big\{E^{\lambda}(v) - \langle v,\ \rho\rangle\Big\}$$

$$F_{\mathrm{LL}}^{\lambda}(\rho) = \inf_{\psi\mapsto\rho}\ \langle\psi|\widehat{T} + \lambda\widehat{W}|\psi\rangle \qquad F_{\mathrm{L}}^{\lambda}(\rho) = \inf_{\Gamma\mapsto\rho}\ \mathrm{Tr}\Big[(\widehat{T} + \lambda\widehat{W})\Gamma\Big]$$

If $\rho \in L^1 \cap L^3$, then $F^{\lambda}(\rho) = F_{\mathrm{L}}^{\lambda}(\rho) \leq F_{\mathrm{LL}}^{\lambda}(\rho)$

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