Quantum Electrodynamic Density-Functional Theory

The Quantum Rabi Model and the Lieb Approach

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Acknowledgements

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Density Functional Theory

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- Prerequisites
- The Hohenberg–Kohn Theorem
- The Lieb Approach

2 **The Quantum Rabi Model**
- Motivation
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The Many-Body Schrödinger Problem

$$\widehat{H}\,|\psi\rangle = E\,|\psi\rangle\,, \quad |\psi\rangle \in \mathcal{H} \tag{1}$$

N particles $\longrightarrow \mathcal{H} = \mathcal{H}_1^{\bigotimes N}$

$$\dim \mathcal{H} = (\dim \mathcal{H}_1)^N$$

Example

Spin-$1/2$ system: $\mathcal{H}_1 = \mathbb{C}^2 \longrightarrow \dim \mathcal{H}_1 = 2$

$$\dim \mathcal{H} = 2^N$$

$N = 40 \longrightarrow \dim \mathcal{H} = 2^{40} \approx 10^{12} \rightsquigarrow$ 16 TB per vector

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The Minimisation Problem

$$E_0 = \min_{|\psi\rangle} \langle\psi|\widehat{H}|\psi\rangle \tag{2}$$

The One-Particle Density

$$\rho(\mathbf{r}) = N \iint \cdots \int |\psi(\mathbf{r}, \mathbf{x}_2, \ldots, \mathbf{x}_N)|^2 \, \mathrm{d}\sigma \, \mathrm{d}\mathbf{x}_2 \, \mathrm{d}\mathbf{x}_3 \ldots \mathrm{d}\mathbf{x}_N \tag{3}$$

The Hohenberg–Kohn Theorem

Given an arbitrary potential $v(\mathbf{r})$, the Schrödinger equation uniquely determines the ground-state density $\rho(\mathbf{r})$[1].

$$v(\mathbf{r}) \longrightarrow \rho(\mathbf{r})$$

Theorem (Hohenberg & Kohn [1])

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

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

[1] up to possibly combinations for degenerate solutions

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The Universal Density Functional

$$E_0(v) = \inf_{\rho} \left[\mathcal{F}(\rho) + \langle v, \rho \rangle\right] \tag{4}$$

$\mathcal{F}(\rho)$ – The density-functional

$$\mathcal{F}(\rho) = \sup_{v} \left[E_0(v) - \langle v, \rho \rangle\right] \tag{5}$$

$\mathcal{F}(\rho)$ – Convex

$E_0(v)$ – Concave

Equations (4) and (5) $\longrightarrow$ Fenchel–Young inequality:

$$E_0(v) - \mathcal{F}(\rho) \leq \langle v, \rho \rangle \tag{6}$$

The Levy–Lieb Functional

Consider the Hamiltonian

$$\widehat{H} = \underbrace{\widehat{T} + \lambda\widehat{W}}_{\text{internal}} + \widehat{V} \tag{7}$$

$$\begin{aligned} \mathcal{F}_{\mathrm{LL}}(\rho) &= \inf_{\psi\to\rho} \langle\psi|\widehat{T} + \lambda\widehat{W}|\psi\rangle \\ &= \langle\psi(\rho)|\widehat{T} + \lambda\widehat{W}|\psi(\rho)\rangle \end{aligned} \tag{8}$$

[2, 3]

The Levy–Lieb Constrained Search

The variational property

$$E_0(v) = \inf_{\substack{\psi \in \mathcal{Q}(\widehat{H}) \\ v \mapsto \widehat{V}}} \langle \psi | \widehat{T} + \lambda \widehat{W} + \widehat{V} | \psi \rangle \tag{9}$$

$$\begin{aligned} E_0(v) &= \inf_{\rho} \left[\mathcal{F}_{\mathrm{LL}}(\rho) + \langle v, \rho \rangle \right] \\ &= \inf_{\rho} \left[\mathcal{F}_{\mathrm{LL}}(\rho) + \int \mathrm{d}\mathbf{r}\, v(\mathbf{r}) \rho(\mathbf{r}) \right] \end{aligned} \tag{10}$$

The Quantum Rabi Model

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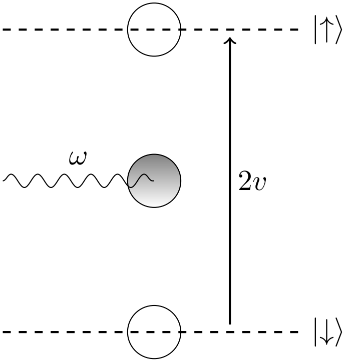
Motivation

- Importance of light-matter interactions
- Simple model
- Describes experimentally accessible coupling strengths [4]
- Study ground state effects of coupling photons to electronic systems
- Studying an (almost) explicit form of a DFT functional
- Investigate Moreau–Yosida regularisation in DFT

The Model

Internal Hamiltonian

$$\widehat{H}_{\text{int}} = \underbrace{-t\widehat{\sigma}_x}_{\text{TLS kin.}} + \overbrace{\lambda\sqrt{2\omega}\widehat{\sigma}_z\widehat{x}}^{\text{TLS-QHO coupling}} + \underbrace{\frac{1}{2}\widehat{p}^2 + \frac{\omega^2}{2}\widehat{x}^2}_{\text{QHO}} \qquad (11)$$



Full Hamiltonian

$$\widehat{H}(v,j) = \widehat{H}_{\text{int}} + v\widehat{\sigma}_z + j\widehat{x} \qquad (12)$$

The Rotating Wave Approximation

Quantum Rabi model – Analytically hard

Standard trick – Rotating wave approx.

$$\widehat{H}_{\mathrm{JC}} = -t\widehat{\sigma}_x + \lambda\left(\widehat{\sigma}_+\widehat{a} + \widehat{\sigma}_-\widehat{a}^\dagger\right) + \omega\widehat{a}^\dagger\widehat{a} + \frac{\omega}{2} \tag{13}$$

$\widehat{\sigma}_\pm = \left(\widehat{i\sigma_y} \pm \widehat{\sigma}_z\right)/2$, ↑ The Jaynes–Cummings model

Symmetries

Quantum Rabi model	Jaynes–Cummings model
Discrete ($\mathbb{Z}_2$)	Continuous ($U(1)$)

[4]

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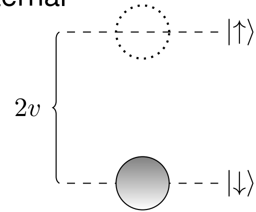
Internal & External Variables

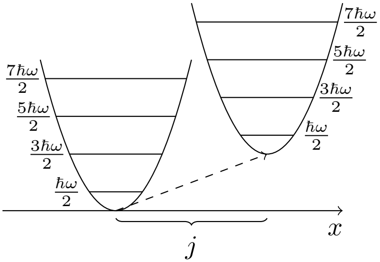
Internal

$$\sigma = \langle\psi|\widehat{\sigma}_z|\psi\rangle = \int_{\mathbb{R}} \mathrm{d}x \left(|\psi_+(x)|^2 - |\psi_-(x)|^2\right) \quad (14)$$

$$\xi = \langle\psi|\widehat{x}|\psi\rangle = \int_{\mathbb{R}} \mathrm{d}x\, x\left(|\psi_+(x)|^2 + |\psi_-(x)|^2\right) \quad (15)$$

External





The Energy Functional

$$\mathcal{W} := \left\{ |\psi\rangle \in \mathcal{H} : \|\psi\|^2 = 1,\ \left\|\widehat{p}^2\psi\right\|^2 < \infty \right\} \tag{16}$$

$$E_0(v, j) := \inf_{|\psi\rangle \in \mathcal{W}} \langle\psi|\widehat{H}(v, j)|\psi\rangle \tag{17}$$

The Levy–Lieb-Functional

$$\mathcal{F}_{\mathrm{LL}}(\sigma, \xi) := \inf_{|\psi\rangle \in \mathcal{Q}(\widehat{H}_0)} \left\{ \langle\psi|\widehat{H}_{\mathrm{int}}|\psi\rangle : \sigma(\psi) = \sigma,\, \xi(\psi) = \xi,\, |\psi\rangle \in \mathcal{W} \right\} \tag{18}$$

$$\mathcal{D} := \left\{ (\sigma, \xi) \in [-1, 1] \times \mathbb{R} : \sigma = \sigma(\psi),\, \xi = \xi(\psi),\, \mathcal{W} \ni |\psi\rangle \in \mathcal{Q}(\widehat{H}_{\mathrm{int}}) \right\} \subset \mathbb{R}^2 \tag{19}$$

$$E_0(v, j) = \inf_{(\sigma,\xi) \in \mathcal{D}} \left[\mathcal{F}_{\mathrm{LL}}(\sigma, \xi) + v\sigma + j\xi\right] \tag{20}$$

A Hohenberg–Kohn Theorem

Theorem (Weak)

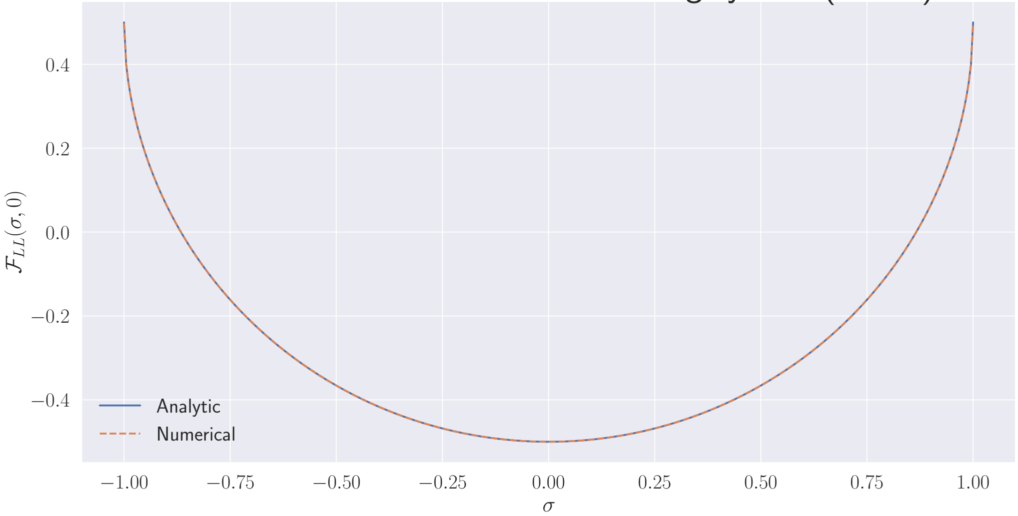
Suppose $|\psi_1\rangle, |\psi_2\rangle \in \mathcal{W}$ *are ground states of* $\widehat{H}(v_1, j_1)$ *and* $\widehat{H}(v_2, j_2)$ *respectively. If both* $|\psi_1\rangle, |\psi_2\rangle \mapsto (\sigma, \xi)$, *then* $|\psi_2\rangle$ *is a ground state of* $\widehat{H}(v_1, j_1)$, *and* $|\psi_1\rangle$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

Theorem (Strong)

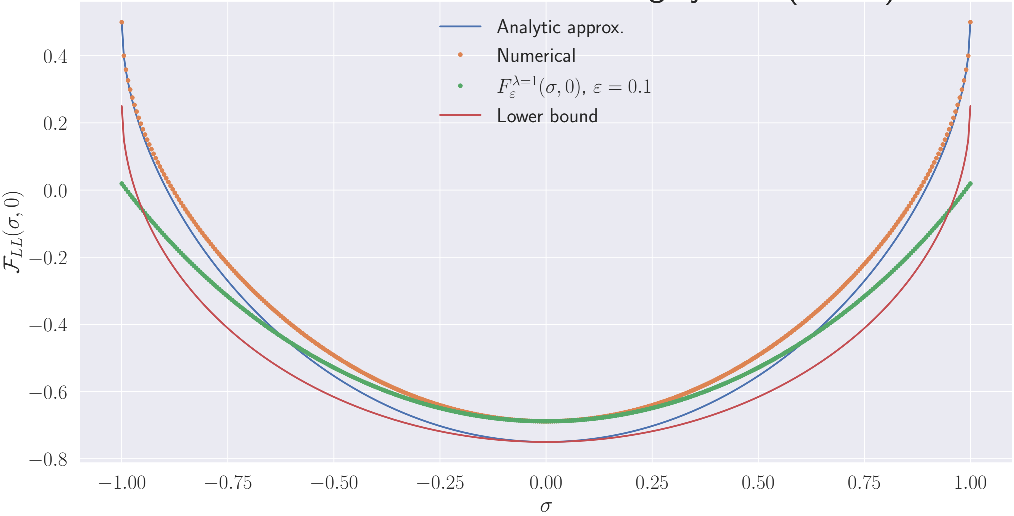
The mapping $(v, j) \mapsto (\sigma, \xi) \in (-1, 1) \times \mathbb{R}$ *is an injection.*

Based on notes by Ruggenthaler [5]

Universal functional - Non-interacting system ($\lambda = 0$)



Universal functional - Interacting system ($\lambda = 1$)



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Moreau–Yosida Regularisation

$\mathcal{F}_{\mathrm{LL}}(\sigma, \xi)$ – much simpler than in standard DFT

Goal: learn more about correlation part of $\mathcal{F}_{\mathrm{LL}}(\sigma, \xi)$

$$\overline{\mathcal{F}}_{\mathsf{LL}}(\sigma, \xi) = \begin{cases} \mathcal{F}_{\mathsf{LL}}(\sigma, \xi), & \forall (\sigma, \xi) \in \mathrm{dom}(\mathcal{F}_{\mathrm{LL}}) \\ +\infty, & \text{otherwise} \end{cases} \tag{21}$$

Moreau–Yosida regularisation:

$$\mathcal{F}^{\varepsilon}_{\mathrm{LL}}(\sigma, \xi) = \inf_{(\rho,\chi)\in\mathbb{R}^2} \left\{ \overline{\mathcal{F}}_{\mathrm{LL}}(\rho, \chi) + \frac{1}{2\varepsilon}\left[(\rho - \sigma)^2 + (\chi - \xi)^2\right] \right\} \tag{22}$$

Many Modes

$$\widehat{H}_{1\gamma} = -t\widehat{\sigma}_x + \lambda\widehat{\sigma}_z\widehat{a} + \omega\left(\widehat{a}^\dagger\widehat{a} + \frac{1}{2}\right) + v\widehat{\sigma}_z + j\widehat{a} + \text{h.c.} \tag{23}$$

$$\widehat{H}_{N\gamma} = -t\widehat{\sigma}_x + v\widehat{\sigma}_z + \lambda\widehat{\sigma}_z\sum_{n=1}^{N}\widehat{a}_n + \sum_{n=1}^{N}\omega_n\left(\widehat{a}_n^\dagger\widehat{a}_n + \frac{1}{2}\right) + \sum_{n=1}^{N}j_n\widehat{a}_n + \text{h.c.} \tag{24}$$

Many Sites – The Dicke Model

$$\widehat{H}_{\text{internal}} = \underbrace{\omega \widehat{a}^\dagger \widehat{a} + \frac{\omega}{2}}_{\text{QHO}} - t \overbrace{\sum_{j=1}^{N} \widehat{\sigma}_{x_j}}^{\text{TLS kin.}} + \underbrace{\frac{\lambda}{\sqrt{N}} \left(\widehat{a} + \widehat{a}^\dagger \right) \sum_{j=1}^{N} \widehat{\sigma}_{z_j}}_{\text{coupling term}} \tag{25}$$

$$\widehat{H} = \widehat{H}_{\text{internal}} + \vec{v} \cdot \vec{\sigma}_z + j \left(\widehat{a} + \widehat{a}^\dagger \right) \tag{26}$$

$$\vec{v} = (v_1, v_2, \ldots, v_N) \qquad \vec{\sigma}_z = (\widehat{\sigma}_{z_1}, \widehat{\sigma}_{z_n}, \ldots, \widehat{\sigma}_{z_N})$$

[6]

Extension to Quantum Electrodynamics

Non-relativistic limit of QED $\longrightarrow$ the Pauli–Fierz Hamiltonian

N electrons, two photon polarisations $\lambda = 1, 2 \longleftarrow$ Coulomb gauge

$$H = \frac{1}{2m} \sum_{j=1}^{N} [\vec{\sigma}_j \cdot (p_j - e(A(q_j) + A_{\text{ext}}(q_j)))]^2 + \sum_{k>j} W(q_k - q_j) \tag{27a}$$

$$+ e \sum_{j=1}^{N} \phi_{\text{ext}}(q_j) + \sum_{\lambda=1,2} \int \mathrm{d}^3 k \, \omega_k a^\dagger_{\lambda,k} a_{\lambda,k} - \int \mathrm{d}^3 q \, j_{\text{ext}}(q) \cdot A(q) \tag{27b}$$

$$[q_j, p_k] = i\delta_{j,k}$$

$$\vec{\sigma}_j = \left(\sigma_{x_j}, \sigma_{y_j}, \sigma_{z_j}\right)$$

$A_{\text{ext}}(q) \longleftarrow$ External vector potential

$\phi_{\text{ext}}(q) \longleftarrow$ External scalar potential

$j_{\text{ext}}(q) \longleftarrow$ External charge current

$$A_\lambda(q) = \int \frac{\mathrm{d}^3 k}{(2\pi)^{3/2}} \frac{\epsilon_\lambda}{\sqrt{2\omega_{\lambda,k}}} \left(a_{\lambda,k} e^{ik\cdot q} + a^\dagger_{\lambda,k} e^{-ik\cdot q}\right)$$

[7, 8]

Coming to arXiv

In the summer of 2024 (hopefully)

Thank you for your attention!

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