UNIVERSIDAD DEL PAIS VASCO

Density-Potential Mapping in the Standard and Quantum Electrodynamical Time-Dependent Density Functional Theory

by

Mehdi Farzanehpour

Supervisors:

Prof. Ilya Tokatly

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A thesis submitted in partial fulfillment for the

degree of Doctor of Philosophy

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"Questioning is the shortest path to learning!"

To be proven in the future.

Abstract

This thesis is devoted to the formulation and implications of the time-dependent density functional theory (TDDFT). The work is divided into two main parts. In the first part we develop rigorous theorems for the density-potential mapping in quantum many-body systems on a lattice. We prove the uniqueness of the TDDFT map and demonstrate that a given density is v-representable if both the initial many-body state and the density satisfy certain well-defined conditions. In particular, we show that for a system evolving from its ground state, any density with a continuous second time derivative is v-representable .

Then the lattice TDDFT formulation is extended to cover a system of interacting lattice electrons strongly coupled to cavity photons. We prove that under some mathematical conditions the electron-photon wave function is a unique functional of the electronic density and the expectation value of the photonic coordinate. Then we further generalize the ground state v-representability theorem to include the ground state of a general lattice electron-photon Hamiltonian.

The second part of this thesis is focused on the applications of the analytic density-potential maps in TDDFT and the current-potential map in the time-dependent current density functional theory (TDCDFT).

We use the analytic lattice map to analyze and quantify the role of non-adiabaticity ("memory effects") in the exchange-correlation functional for describing non-linear dynamics of many-body systems. Studying time-dependent resonant processes using the available functional in TDDFT is a big challenge and that is due to their strong non-linear and non-adiabatic character. Here we study the Rabi oscillations within the solvable 2-site Hubbard model as an example for the resonant processes. We construct the exact adiabatic exchange-correlation functional and show that it cannot reproduce resonant Rabi dynamics correctly. It turns out the non-adiabatic contribution to the exact exchange-correlation potential is significant throughout the dynamics and it is small only when the ground state population is significant. Afterwards we reconstruct the exact time-dependent exchange-correlation functional within the two-level approximation. This fully non-adiabatic and explicit density functional captures Rabi dynamics both for resonant and detuned oscillations.

Finally we apply the analytic current-potential maps of TDCDFT in a completely different context. We use them to inverse engineer analytically solvable time-dependent quantum problems. In this approach the driving potential (the control signal) and the corresponding solution of the Schrödinger equation are analytically parametrized in terms of the basic TD(C)DFT observables.

We describe the general reconstruction strategy and illustrate it with few explicit examples ranging from real space one-particle dynamics to controlling quantum dynamics in

a discrete space. In particular we construct a time-dependent potential which generates prescribed dynamics on a tight-binding chain. We also apply our method to the dynamics of spin-1/2 driven by a time-dependent magnetic field and construct an analytic control pulse for the quantum NOT gate.

Acknowledgements

List of Publications

- **Dynamics of observables and exactly solvable quantum problems: Using time-dependent density functional theory to control quantum systems**
  Mehdi Farzanehpour, I. V. Tokatly, arXiv:1506.08786 (2015).

- **Quantum electrodynamical time-dependent density-functional theory for many-electron systems on a lattice**
  Mehdi Farzanehpour, I. V. Tokatly, Physical Review B 90, 195149 (2014).

- **The time-dependent exchange-correlation functional for a Hubbard dimer: quantifying non-adiabatic effect**
  Johanna I. Fuks, Mehdi Farzanehpour, Ilya V. Tokatly, Heiko Appel, Stefan Kurth, Angel Rubio, Physical Review A 88, 062512 (2013).

- **Time-dependent density functional theory on a lattice**
  Mehdi Farzanehpour, I. V. Tokatly, Physical Review B 86, 125130 (2012).

Contents

List of Figures

Abbreviations

DFT   **D**ensity **F**unctional **T**heory

TDDFT   **T**ime-**D**ependent **D**ensity **F**unctional **T**heory

TDCDFT   **T**ime-**D**ependent **C**urrent **D**ensity **F**unctional **T**heory

XC   **E**xchange **C**orrelation

HXC   **H**artree **E**xchange **C**orrelation

KS   **K**ohn-**S**ham

NLSE   **N**on-**L**inear **S**chrödinger **E**quation

QED   **Q**uantum **E**lectro**D**ynamics

ODE   **O**rdinary **D**ifferential **E**quation

PDE   **P**artial **D**ifferential **E**quation

Chapter 1

Introduction

The physical and chemical properties of systems composed of low energy particles are determined by the non-relativistic Schrödinger equation . This equation has been proven extremely successful (essentially exact) in describing quantum mechanical systems, e.g. atoms and molecules, but unfortunately it can be solved efficiently only for systems with very few interacting particles.

The Schrödinger equation for a system of N interacting particles in a three dimensional space is a partial differential equation with $3N$ special coordinates which are mutually coupled through the Coulomb interaction. This makes the complexity of the wave function to increase exponentially with the number of particles until eventually we encounter an "exponential wall" .[1] To put it in perspective, to discretize the space even for a "humble" system with around 20 electrons (For example, a water molecule, or a Titanium atom),the number of bits to store the wave function can easily exceed the estimated number of atoms in the observable universe $10^{72} - 10^{82}$. [1]

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[1]To make this fact more clear let's make crude estimation of amount of memory occupied by the discretized wave function. For the discretization we define a mesh with M points in each direction. by choosing the proper basis set for this discrete space we can expand the state of the system

$$|\Psi(t)\rangle = \sum_{\mathbf{r}_1, \mathbf{r}_2, ..., \mathbf{r}_N} \psi(\mathbf{r}_1, \mathbf{r}_2, ..., \mathbf{r}_N; t) |\mathbf{r}_1\rangle|\mathbf{r}_2\rangle \cdots |\mathbf{r}_N\rangle, \tag{1.1}$$

where $\mathbf{r}_i = (x_i, y_i, z_i)$ is the position of the ith electron which takes values on discrete points. $\psi(\mathbf{r}_1, \mathbf{r}_2, ..., \mathbf{r}_N; t)$ is the interpretation of the wave function in a discrete space. In total there are M^{3N} ψ's and if each occupies a bits we will need the total memory R of

$$R = aM^{3N}. \tag{1.2}$$

So if we just decide to choose $M = 10$, 1000 mesh point in total, to store the state of a caffeine molecule,$C_8H_{10}N_4O_2$, with 102 electrons we need at least 5×10^{287} TB. This is considering each ψ only takes $x = 4$ bits to store. To put things in perspective, it is around 5.5×10^{281} times the estimated

However there is an ingenious and extremely nontrivial way to treat this problem and that is by using the one-to-one correspondence between single particle density and the external potential in the Density Functional Theory (DFT) and its time-dependent version (TDDFT).

DFT is a formulation of the stationary quantum mechanics derived from the time-independent Schrödinger equation which states that the ground state density of many-body quantum system is uniquely determined by the external potential.[2] In other words, there is a unique density for a given external potential which minimizes the total energy functional of the system. This means that to calculate any observable in the ground state, in principle, all we need is the ground state density and, thus, the complicated many-body wave function of the ground state does not contain any information in addition to that in the single particle ground state density.

The pinnacle of this theory is the Kohn-Sham construction which makes DFT usable in practice.[3] By definition the Kohn-Sham system is an auxiliary system of N non-interacting electrons in an effective external potential which has the same ground density as the original one with N interacting electrons. The effective, Kohn-Sham, potential is a unique functional of the density and this reduces the static many-body problem to a self consistent Hartree-like problem. [2]

Time-dependent density functional theory (TDDFT) is a non-trivial extension of DFT to non-stationary systems in the presence of time-dependent external potentials. This theory is rapidly becoming the method of choice for modeling dynamics of the realistic many-body systems. The reasons for the popularity of TDDFT are the same as those for the ground state DFT. It significantly reduces complexity of the problem by allowing to calculate the density through an auxiliary system of non-interacting Kohn-Sham particles. The possibility of such a reduction rests on two fundamental mathematical statements: (i) the one-to-one map between the density and the external potential and (ii) v-representability of the density both in the interacting and non-interacting system, i. e. the time-dependent density can be as a result of a time evolution driven by an external scalar potential. The first statement, known as a mapping theorem, guarantees that the many-body wave function and, thus, any observable are unique functionals of the

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size of the world wide web (Internet) at 2010 and 10^{205} times the number of atoms in the observable universe.

[2] An explicit definition of the Kohn-Sham system is given in chapter 2.

density. The v-representability is required for both the interacting and non-interacting systems to guarantee the Kohn-Sham construction.

Finding a rigorous proof for the v-representability and uniqueness of the map is a highly nontrivial and, to some extend, still an open problem. Moreover, having such a proof of the concept does not necessarily translate to a practical use unless we either discover the universal map explicitly, which is highly unlikely, or develop a proper approximation which captures properties of the system. More on the latter will come later.

The first mapping theorem for the static DFT was presented in a paper by Hoheberg and Kohn [2] while a general solution of the more tricky v-representability problem appeared much later and only for lattice systems. [4, 5]

Proving the corresponding theorems in TDDFT turned out to be even more difficult because of the absence of minimum principle for the dynamics. Thus one has to use equations of motion to prove any mappings statement. In fact, only 20 years after the first paper on DFT, Runge and Gross (RG) succeeded to find a sufficiently general proof of the TDDFT mapping theorem for a class of analytic in time (t-analytic) potentials [6].

More than a decade after an attempt to attack the more tricky problem of time-dependent v-representability has been performed in Ref. [7] by assuming t-analyticity both for potentials and for allowed densities. Under this restriction a formal power series for the potential can be uniquely reconstructed from a given Taylor expansion of the density. Unfortunately the convergence of that series is not proven up to now, and thus a complete solution of the v-representability problem within the series expansion technique is still missing. We note that the issues of t-analyticity and a uniform convergence of power series in quantum dynamics are not as exotic as it may appear at the first sight [8–10]. Despite a number of indications[11, 12] that t-analyticity was not a fundamental limitation of the theory, a question of a more general and clean justification of TDDFT remained open for many years.

In the past few years it has been recognized [9, 13–18] that the existence of all TDDFT-type theories is equivalent to the solvability of a certain universal nonlinear many-body problem which determines the potential and the many-body wave function in terms of a given basic observable. Mathematically this universal problem can be posed in

two different forms. The first possibility is to view it as a Cauchy problem for a special nonlinear Schrödinger equation (NLSE) [9, 13–16]. The uniqueness and the existence of solutions to this NLSE are equivalent, respectively, to the mapping and the v-representability problems in TDDFT. Alternatively it can be formulated as a fixed point problem for a certain nonlinear map in the space of potentials [17, 18]. First applications of the two formulations above appeared almost simultaneously in Refs. [16] and [17].

The fixed point approach has been used in Ref. [17] to address the existence of the "classical" TDDFT in its original Runge-Gross form. In this work the t-analyticity requirement was relaxed and effectively replaced by a more physical (though still unproven) assumption – a boundness of a certain generalized response function related to a stress-density correlator.

A rigorous formulation of a time-dependent current density functional theory (TD-CDFT) on a lattice was presented in Ref. [16] within NLSE formulation of the problem. The lattice TDCDFT turned out to be the first and, in fact, the simplest example of a TDDFT-type theory for which both the mapping and the v-representability theorems have been proven without any unjustified assumption.

However despite all the efforts, a rigorous proof of the TDDFT formalism is yet to be presented. Moreover, in the most general setting the v-representability problem remains open till now. In chapter 3 we answer the fundamental questions of TDDFT by reformulating the density-potential map for a lattice. We realize that this reduction massively simplifies the mathematical structure of the problem. We further develop the lattice NLSE technique of Ref. [16] to address a long standing problem of TDDFT for lattice many-body systems. It may seem surprising, but before this work even the Runge-Gross mapping theorem for t-analytic potentials for the lattice TDDFT was absent. This should be contrasted to the lattice TDCDFT where the standard power series argumentation can be easily adapted [19]. Many discussions of technical and conceptual difficulties of the lattice TDDFT can be found in the literature [20–24]. We demonstrate that the lattice analogue of NLSE answers to those conceptual problems. We prove both the uniqueness and existence theorems for the lattice TDDFT and analyze conditions which have to be imposed on the initial state and the density to guarantee v-representability. In particular we demonstrate that practically any properly normalized

density is locally *v*-representable provided the dynamics start from the ground state. These results as the first rigorous proof of the TDDFT formalism put applications of TDDFT to various lattice models [20–24, 24–28] on a firm ground, and shed new light on the general mathematical structure of TDDFT.

Afterwards we generalize the lattice TDDFT to include lattices strongly coupled to the quantum light. This is in contrast to the standard TDDFT which assumes the system is driven by a classical time-dependent electric field. Although the TDDFT approach is sufficient for most typical situations in quantum chemistry and condensed matter physics, however, in the recent years, with the impressive progress in the fields of cavity and circuit quantum electrodynamics (QED) it has been made possible to experimentally study systems interacting strongly with quantum light, like atoms in optical cavity [29–31], superconducting qubits and quantum dots [32–35] , trapped ions [36] and molecules interacting with cavity photons. [37–39].

Recently a generalization of TDDFT mapping theorem for quantum many-electron systems coupled to cavity photons has been proposed [40, 41]. This theory, called QED-TDDFT, relies on the statement that there exists a unique map from a set of basic variables, the density and the expectation value of the Bosonic field, to the set of driving potentials. The latter consists of a classical external electromagnetic field driving the electronic subsystem and an external radiative source, e.g. external current, stimulating the photonic modes. In Ref. [40] the uniqueness of this generalized mapping has been demonstrated using the Taylor expansion technique under the previously discussed *t*-analyticity assumption. [6] However, as we discussed, the Taylor expandability is not a natural criteria deduced from the equations of motion. In addition the *v*-representability problem, convergence of the Taylor series, is yet to be addressed.

In chapter 4 we extend the NLSE formalism of chapter 3 to cover the situation where the lattice electrons are interacting strongly with quantized photonic modes. First we demonstrate the procedure by proving the QED-TDDFT map for the simplest nontrivial system of an electron on a two-site lattice (a Hubbard dimer) coupled to a single photonic mode. It is worth noting that indeed, the QED-TDDFT formulation for a dimer coupled to a quantum Bose field has its own value. This system is equivalent to well known and popular models as the quantum Rabi model and the spin-boson model [42–44] which have a wide variety of applications ranging from quantum optics [45] and molecular physics

[46] to the magnetic resonance in solid state physics. [47] We prove that, provided some well-defined conditions are fulfilled, there exists a unique mapping from the set of basic variables to the driving potentials. Then we extend the QED-TDDFT mapping theorem to the general case of N interacting electrons on an M-site lattice coupled to multiple photonic modes. We also prove that, similar to the standard lattice TDDFT, the local existence/v-representability is guaranteed if the dynamics start from the ground state of a lattice Hamiltonian. These rigorous statements answer both the v-representability and uniqueness problems of QED-TDDFT for lattices.

Next, we move our attention to the direct consequence of the TDDFT and that is the functional dependence of the external potential on the density. As a result of the TDDFT mapping provided a density is non-interacting v-representable , its dynamics can be reproduced by an auxiliary system of non-interacting Kohn-Sham particles with a unique effective Kohn-Sham potential. Traditionally the Kohn-Sham potential is expressed as a sum of three terms with each having a particular physical significance. The first term is the external potential of the interacting system and the second term, Hartree potential, is a mean-field Coulomb interaction through the density. The remaining potential from the interaction is put in a term called exchange-correlation potential.

However, the functional dependence of the exchange-correlation potential on the density, except for very special occasions, is unknown and most probably never be known. So not surprisingly, it has been an ever continuing quest to better approximating it .[48, 49] The TDDFT formalism, with the available approximations to the exchange-correlation functional, has been greatly successful in describing optical properties of a large variety of molecules and nanostructures. [50–52] However these approximations exhibit serious deficiencies in the description of non-linear processes, long range charge transfer [53–55] and double excitations [56–58], to mention a few.

The theoretical challenge is to improve the available functionals in order to capture the nonlocality both in space and time of the exact exchange-correlation functional which depends on the history of the density, the initial (interacting) many-body state and the initial Kohn-Sham state.[59–62] Currently almost all of the TDDFT calculations are done under the adiabatic approximation which assumes the instantaneous density is the ground state of density of the system. This approximation is completely local in time and neglects both the history and the initial state dependence of the exact functional.

In chapter 5 we exemplify the failure of the adiabatic approximation of the exchange-correlation functional in reproduction of the non-linear dynamics for a solvable lattice model, 2-site Hubbard model.[63–65] Specifically, we study resonant and slightly de-tuned resonance Rabi oscillations where the population of states changes dramatically in time. We first derive the exact ground state Hartree-exchange-correlation (Hxc) functional for the Hubbard dimer using the Levy-Lieb constrained search. [66–68] Then the functional is used in a TDDFT context with the instantaneous time-dependent density as the input which constitutes the exact adiabatic approximation. By carefully studying and quantifying the dynamics produced by TDDFT with the adiabatic Hxc potential we demonstrate that it fails both quantitatively and qualitatively to describe Rabi oscillations. In addition by comparing it with the exact Hxc functional calculated from the propagated density we identify the source of the failure. Afterwards we apply the analytic density-potential map for lattice systems of chapter 3 to derive an explicit, fully non-adiabatic exchange-correlation density functional which correctly captures all features of Rabi dynamics in the Hubbard dimer. This functional is considered one of the main results of chapter 5.

Another potential application of the TD(C)DFT functionals, proposed in this thesis, is a reconstruction/inverse engineering analytic control pulses for single-particle quantum dynamics. Analytically solvable quantum problems have been always of interest in the theoretical physics since they deepen our intuition of the quantum mechanical systems. However unlike the static quantum problems where there is a handful of solvable examples [69], there is just a very few analytically solvable examples with a time-dependent potential. To name a few: solutions of the Landau-Zener [70, 71] and Rabi [72] problems, or the solution for a driven harmonic oscillator [73, 74] which is closely related to a so-called harmonic potential theorem.[75–77]

In the recent years and with the impressive progress in the fields of quantum computing and quantum control the interest in analytically solvable quantum dynamical problems is renewed. In order to further develop quantum gates it is necessary to accurately control [78–80] and prepare the state of a qubit [81–83]. It has been recognized that analytic pulses in quantum control lead to a more robust evolution against errors and noise in pulse parameters [78, 84, 84–87] which can explain the great interest in finding new pulses to analytically control two-level systems. [88–96]

In the last decade a new trend has emerged where they inverse engineer time-dependent Hamiltonian to result prescribed dynamics for a set of parameters. Most of the examples of these studies are based on two level systems [94, 97–100] and a few for the three level systems. [101]

In chapter 6 we propose an alternative strategy of reconstructing time-dependent driving potentials for analytically solvable quantum problems. Our proposal employs the explicit (current) density-(vector) potential map of TD(C)DFT for single particle systems.[102–106] These maps imply that the knowledge of some properly chosen one particle observables, such as the density or the current, is sufficient to explicitly reconstruct the corresponding driving fields, and therefore, the full wave function of the system. The same strategy would be applicable to a general many-particle quantum system, if a proper reconstruction of the density-potential map is possible. For example, recently using the global fixed point formalism, mentioned above, the external potential for a prescribed density evolution in a model system was numerically constructed. [107, 108]

In chapter 6 we address, within a common scheme, control problems for the real space dynamics and for dynamics of discrete systems with a finite dimensional Hilbert space, such as a motion of quantum particle on tight-binding lattices, or the dynamics of a spin in the presence of a time-dependent magnetic field. To illustrate our strategy of inverse engineering we will recover the known exact solution for a driven harmonic oscillator [73, 74], and present nontrivial examples of analytic control for a particle on a finite 1D chain and for a spin-1/2 (qubit) system.

Finally we conclude this work with a short discussion on the results and outlook of the further research.

Chapter 2

Theoretical Background

In this chapter we present some theoretical basics underlying our work. As mentioned in the introduction, the focus of this work is on the mapping theorems for the non-stationary quantum problems. However we think, mainly for two reasons, it makes sense to start with the original mapping theorems for the static problem. First, it reviews the very first steps of a revolution in the computational physics and chemistry which made possible studies of realistic many-electron systems. Second and more important for our work, it emphasizes on the fact that the DFT formalism relies on the minimum principle. This is in contrast to the dynamics where there is no minimum principle to rely and thus the mapping theorems need to be based on the equations of motion.

Then in the section for TDDFT we present a detailed review of the Runge-Gross theorem which proves the uniqueness of the density-potential map. The proof is for Taylor expandable densities and moreover, the v-representability of the density is taken for granted. Later in chapter 3 we see that both of these assumptions can be relaxed on lattices.

Afterwards, we present an alternative formulation of the density-potential mapping problem. This formulation as a mathematically well posed problem, in principle, can lead us to the solution for both v-representability and uniqueness problems. Then a review of the two approaches to this formulation is presented.

This covers most of the basics for chapters 3, 5 and 6. In the last section we present a derivation for the electron-photon Hamiltonian in the context of non-relativistic QED.

This provides us with a sufficient material for the chapter 4 where we prove the lattice QED-TDDFT mapping.

2.1 Density functional theory

We start this section with the original Hohenberg-Kohn theorem and its proof. Then we will move onto the Kohn-Sham formulation.

2.1.1 Hohenberg-Kohn Theorem

Let's assume a system of N interacting fermions. The solution to the time-independent Schrödinger equation determines the energy spectrum and eigenstates of the system

$$\hat{H}|\Psi\rangle = E|\Psi\rangle. \tag{2.1}$$

The Hamiltonian $\hat{H}$ is the total energy operator of the system

$$\hat{H} = \hat{T} + \hat{W} + \hat{V}_{ext}, \tag{2.2}$$

where $\hat{T}$ is the kinetic energy operator, $\hat{W}$ the two-particle interaction operator

$$\hat{T} = -\frac{1}{2}\sum_{i=1}^{N}\nabla_i^2, \tag{2.3}$$

$$\hat{W} = \frac{1}{2}\sum_{i\neq j}w(\mathbf{r}_i,\mathbf{r}_j). \tag{2.4}$$

$\hat{V}_{ext}$, the time-independent external scalar potential, is a multiplicative operator which includes the nuclei potential in the case of Born-Oppenheimer approximation

$$\hat{V}_{ext} = \sum_{i=1}^{N}v(\mathbf{r}_i). \tag{2.5}$$

The density operator of an N-particle system is defined

$$\hat{n}(\mathbf{r}) = \sum_{i=1}^{N}\delta(\mathbf{r}-\mathbf{r}_i). \tag{2.6}$$

Considering the Hamiltonian (2.2) to have a non-degenerate ground state $|\Psi_g\rangle$, with the eigenenergy E_g, the expectation value of the density operator at the ground state $n_g(\mathbf{r})$ is given by

$$n_g(\mathbf{r}) = N \int d\mathbf{r}_2 \cdots \int d\mathbf{r}_N |\psi_g(\mathbf{r}, \mathbf{r}_2, ..., \mathbf{r}_N)|^2, \tag{2.7}$$

where the ground state wave function $\psi_g(\mathbf{r}_1, \mathbf{r}_2, ..., \mathbf{r}_N) = \langle \mathbf{r}_1, \mathbf{r}_2, ..., \mathbf{r}_N | \Psi_g \rangle$.

As a result of Eqs. (2.1) and (2.7) we conclude that the ground state $|\Psi_g\rangle$ and the ground state density n_g are functionals of the external potential, but the question in DFT is if these maps are unique and thus invertible? The Hohenberg-Kohn theorem is an attempt to answer this question.

Theorem 2.1.1 (Hohenberg-Kohn Theorem). In a finite interacting system of N fermions with a given interaction $\hat{W}$ the map from the external potential $\hat{V}_{ext}$ to the ground state density n_g is unique, up to a constant shift in the potential.

Proof. We prove the theorem in a two step sequence and both using the proof by contradiction. In the first step we prove that the map from the external potential to the ground state wave function is unique, $\{V_{ext}\} \leftrightarrow \{\Psi_g\}$. Then we prove the uniqueness statement for the map from the ground state wave function to the ground state density, $\{\Psi_g\} \leftrightarrow \{n_g\}$.

To prove the first statement we show two external potentials, $\hat{V}_{ext}$ and $\hat{V}'_{ext}$, which differ by more than a constant, $\hat{V}_{ext} \neq \hat{V}'_{ext} + C$ cannot lead to the same ground state $|\Psi_g\rangle$. So, for the moment, let assume that two potentials $\hat{V}_{ext}$ and $\hat{V}'_{ext}$ have the same ground states $|\Psi_g\rangle = |\Psi'_g\rangle$. Therefore writing two eigenvalue equations for $\hat{H} = \hat{T} + \hat{W} + \hat{V}$ and $\hat{H}' = \hat{T} + \hat{W} + \hat{V}'$

$$\hat{H}|\Psi_g\rangle = E_g|\Psi_g\rangle, \tag{2.8a}$$

$$\hat{H}'|\Psi'_g\rangle = E'_g|\Psi'_g\rangle, \tag{2.8b}$$

and then subtracting them we get

$$\left(\hat{V}_{ext} - \hat{V}'_{ext}\right)|\Psi_g\rangle = \left(E_g - E'_g\right)|\Psi_g\rangle. \tag{2.9}$$

Since the external potential is a multiplicative operator we can divide both sides by $|\Psi_g\rangle$ provided that the ground state is non-zero for a set of positive measure.[109] This results

$$\hat{V}_{ext} - \hat{V}'_{ext} = E_g - E'_g, \tag{2.10}$$

which is in contradiction with the assumption on $\hat{V}_{ext}$ and $\hat{V}'_{ext}$.

Like the first step, we prove the second one by contradiction. We prove that if two different ground states, $|\Psi_g\rangle \neq e^{i\beta}|\Psi'_g\rangle$, have the same density n_g we encounter a contradiction.

First let show that the expectation value of the Hamiltonian $\hat{H}$ with respect to $|\Psi'_g\rangle$ is always bigger than the ground state energy of $\hat{H}$. For the expectation value of $\hat{H}$ we have

$$\langle \Psi'_g|\hat{H}|\Psi'_g\rangle = \sum_{n=0} |a_n|^2 E_n, \tag{2.11}$$

where we expanded $|\Psi'_g\rangle$ over the eigenbasis of $\hat{H}$, $|\Psi'_g\rangle = \sum_{n=0} a_n|\Psi_n\rangle$, with $n = 0$ representing the ground state. On the other hand for the summation on the right hand side of the equation we have:

$$\sum_{n=0} |a_n|^2 E_n > E_g \sum_{n=0} |a_n|^2 = E_g. \tag{2.12}$$

Thus we find:

$$E_g < \langle \Psi'_g|\hat{H}|\Psi'_g\rangle. \tag{2.13}$$

As we see later this inequality, known as the minimum energy principle, is the essential statement in the Hohenberg-Kohn this theorem and in general DFT.

For the expectation value on the right hand side of the equation we can write

$$\langle \Psi'_g|\hat{H}|\Psi'_g\rangle = \langle \Psi'_g|\hat{H} - V'_{ext} + V_{ext}|\Psi'_g\rangle = E'_g + \int d\mathbf{r} \left(v'(\mathbf{r}) - v(\mathbf{r}) \right) n_g(\mathbf{r}), \tag{2.14}$$

where we used he definition of the external potential Eq. (2.31). Then we substitute the expectation value back into the inequality (2.13)

$$E_g < E'_g + \int d\mathbf{r} \left(v'_{ext}(\mathbf{r}) - v_{ext}(\mathbf{r}) \right) n_g(\mathbf{r}). \tag{2.15}$$

We can also write the corresponding inequality for E'_g

$$E'_g < E_g + \int d\mathbf{r} \left(v_{ext}(\mathbf{r}) - v'_{ext}(\mathbf{r}) \right) n_g(\mathbf{r}). \tag{2.16}$$

Finally adding the two inequalities (2.15) and (2.16) we get

$$E_g + E'_g < E_g + E'_g, \tag{2.17}$$

which is an obvious contradiction. Therefore the map from the ground state wave function to the ground state density is unique and thus bijective, $\{\Psi_g\} \leftrightarrow \{n_g\}$.

Consequently the map from the external potential to the ground state density is unique, $\{V_{ext}\} \leftrightarrow \{n_g\}$. $\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\square$

The main argument in the Hohenberg-Kohn proof, the minimum principle (2.13), underlies the density-potential map in DFT. As a direct consequence of this map, the expectation value of any observable $\hat{O}$ in the ground state is uniquely determined by the ground state density n_g. Mathematically speaking, the ground state expectation value of $\hat{O}$ is a unique functional of n_g

$$O[n_g] = \langle \Psi_g[n_g] | \hat{O} | \Psi_g[n_g] \rangle. \tag{2.18}$$

One of these observables is the total ground state energy of the system, the expectation value of the total Hamiltonian (2.2) with respect to $|\Psi_g\rangle$

$$E_{v_{ext}}[n_g] := \langle \Psi_g[n_g] | \hat{T} + \hat{W} + \hat{V}_{ext} | \Psi_g[n_g] \rangle = F_{HK} + \int d\mathbf{r} v(\mathbf{r}) n_g(\mathbf{r}), \tag{2.19}$$

where F_{HK} is the Hohenberg-Kohn energy functional which is sum of the kinetic and interaction energies.

As a consequence of the minimum energy principle (2.13), the energy in the right hand side is minimum if only the density is the ground state density of the Hamiltonian with the specified $\hat{V}_{ext}$ and therefore the ground state density can be found through a variational search: [66, 110, 111]

$$E_g = \min_{n \in \mathcal{N}} E_v[n]. \tag{2.20}$$

where $\mathcal{N}$ is the set of the v-representable ground state densities. One can also rewrite the variational principle in a differential form

$$\frac{\delta}{\delta n(\mathbf{r})} E_v[n]\bigg|_{n=n_g} = 0. \tag{2.21}$$

However one needs to show first, there is a proper norm convergence in $\mathcal{N}$ in the vicinity of the ground state density n_g and, second, the energy functional is differentiable.

2.1.2 Kohn-Sham system in DFT

Lets assume an auxiliary system of N non-interacting fermions described by the Hamiltonian

$$\hat{H}_s = \hat{T} + \hat{V}_s. \tag{2.22}$$

According to the Hohenberg-Kohn , the ground state energy of a non-interacting system is a unique functional of its ground state density n_s. Therefore from (2.19) we have:

$$E_{v_s}[n_s] = T_s[n_s] + \int d\mathbf{r} v_s(\mathbf{r}) n_s(\mathbf{r}), \tag{2.23}$$

where $T_s[n]$ is the universal kinetic energy functional.

This non-interacting system is Kohn-Sham if it reproduces the same ground state density of the interacting system under the study

$$n_s(\mathbf{r}) = n_g(\mathbf{r}). \tag{2.24}$$

Obviously this is possible if the ground state density of the interacting system is also non-interacting v-representable .

From the Hohenberg-Kohn theorem, again, we know that the ground state wave function is uniquely determined by the external potential for a specified interaction. In this case since the particles are non-interacting the non-degenerate ground state can be written

in terms of a single Slater determinant

$$\psi_s(\mathbf{r}_1, \mathbf{r}_2, ..., \mathbf{r}_N) = \frac{1}{\sqrt{N!}} \begin{vmatrix} \varphi_1(\mathbf{r}_1) & \varphi_2(\mathbf{r}_1) & \cdots & \varphi_N(\mathbf{r}_1) \\ \varphi_1(\mathbf{r}_2) & \varphi_2(\mathbf{r}_2) & \cdots & \varphi_N(\mathbf{r}_2) \\ \vdots & \vdots & \ddots & \vdots \\ \varphi_1(\mathbf{r}_N) & \varphi_2(\mathbf{r}_N) & \cdots & \varphi_N(\mathbf{r}_N) \end{vmatrix}, \qquad (2.25)$$

where $\varphi_j(\mathbf{r})$ is the jth eigenfunction of the single-particle Schrödinger equation with the eigenenergy ϵ_j

$$\left(-\frac{\nabla^2}{2} + v_s(\mathbf{r}) \right) \varphi_j(\mathbf{r}) = \epsilon_j \varphi_j(\mathbf{r}). \qquad (2.26)$$

The single particle eigenfunctions, $\varphi_j(\mathbf{r})$, are called the Kohn-Sham orbitals.

Therefore the Kohn-Sham density is a direct sum of the individual orbital densities

$$n_s(\mathbf{r}) = \sum_{j=1}^{N} |\varphi_j(\mathbf{r})|^2. \qquad (2.27)$$

We can rewrite the Kohn-Sham potential by addition and subtraction of the interacting external and Hartree potential

$$v_s[n_s](\mathbf{r}) = v_{ext}(\mathbf{r}) + \int d\mathbf{r} w(\mathbf{r}, \mathbf{r}') n_s(\mathbf{r}) + v_{xc}[n_s](\mathbf{r}) \qquad (2.28)$$

where v_{xc}, the exchange-correlation potential, is a functional of the density and in principle, it includes all many-body effects like correlations and exchange.

Provided the exact exchange-correlation functional is known, the self-consistent solution of the Kohn-Sham equation (2.26) together with Eqs. (2.27) and (2.28) determines the exact ground state density of the Kohn-Sham system and consequently the ground state density of the interacting system. Unsurprisingly, the functional is not known and approximations are needed to be performed. However the DFT theory does not suggest any systematic way to construct approximations which can be strategically improved.

In chapter 5 we find the exact Hohenberg-Kohn functional for a two site Hubbard model by using the constrain search.

2.2 Time-dependent density functional theory

In this section we summarize some of the basic theorems in TDDFT. This theory, in some sense, is a generalization of the static DFT to time-dependent many-particle quantum problems. However, the similarity does not go beyond the very basic idea of density-potential mapping. For example it is not possible to simply extend the DFT's basic theorems to the time-dependent problems.

The system under the study, here, is a group of N interacting identical Fermions, for example electrons, which evolves from a some given initial state $|\Psi_0\rangle$. The time-dependent state $|\Psi(t)\rangle$ is the solution to the time-dependent many-body Schrödinger equation

$$i\partial_t|\Psi(t)\rangle = \hat{H}(t)|\Psi(t)\rangle. \tag{2.29}$$

The system is characterized by a non-relativistic time-dependent Hamiltonian

$$\hat{H}(t) = \hat{T} + \hat{W} + \hat{V}_{ext}(t), \tag{2.30}$$

where the kinetic energy $\hat{T}$ and the two-particle interaction are given by Eqs. (2.3) and (2.4) and the external potential $\hat{V}_{ext}$ is a classical time-dependent scalar potential

$$\hat{V}_{ext}(t) = \sum_{i=1}^{N} v(\mathbf{r}_i, t). \tag{2.31}$$

The time-dependent density $n(\mathbf{r}, t)$ is the expectation value of the density operator with respect to $|\Psi(t)\rangle$

$$n(\mathbf{r}, t) = N \int d\mathbf{r}_2 \cdots \int d\mathbf{r}_N |\psi(\mathbf{r}, \mathbf{r}_2 \cdots \mathbf{r}_N; t)|^2 \tag{2.32}$$

where, as before, the wave function is the projection of the state vector over the position coordinate $\psi(\mathbf{r}_1, \mathbf{r}_2, ..., \mathbf{r}_N; t) = \langle \mathbf{r}_1, \mathbf{r}_2, ..., \mathbf{r}_N|\Psi(t)\rangle$.

Using the Schrödinger equation (2.29) one can show that expectation value of observables obeys the Heisenberg equation of motion

$$\frac{d}{dt}\langle \hat{O} \rangle = i\langle \Psi(t)|[\hat{H}(t), \hat{O}]|\Psi(t)\rangle + \langle \frac{\partial}{\partial t}\hat{O} \rangle. \tag{2.33}$$

So for the time evolution of the density this gives

$$\frac{d}{dt}n(\mathbf{r}, t) = i\langle\Psi(t)|[\hat{H}(t), \hat{n}(\mathbf{r})]|\Psi(t)\rangle. \tag{2.34}$$

The commutator between $\hat{n}$ and $\hat{W}_{ext}$ and between $\hat{n}$ and $\hat{V}_{ext}$ are zero. and $[\hat{T}, n(\mathbf{r})]$ can also be computed with few lines of algebra. The result is the well-known continuity equation which connects the first derivative of the density $\dot{n}$ to the current density [1]

$$\dot{n}(\mathbf{r}, t) = -\nabla \cdot \mathbf{j}(\mathbf{r}, t), \tag{2.35}$$

where the current density operator is defined $\hat{\mathbf{j}}(\mathbf{r})$

$$\hat{\mathbf{j}}(\mathbf{r}) = -\frac{i}{2}\sum_{j=1}^{N}[\nabla_j\delta(\mathbf{r} - \mathbf{r}_j) + \delta(\mathbf{r} - \mathbf{r}_j)\nabla_j]. \tag{2.36}$$

The continuity equation expresses the conservation of particles, that is the number of particles in a given volume changes only if there is a current flow through its boundary.

2.2.1 The Runge-Gross theorem

The "direct" map from the time-dependent external potential $\hat{V}_{ext}$ to the time-dependent state $|\Psi(t)\rangle$ is defined through the Schrödinger equation (2.29). And the map from the wave function $|\Psi(t)\rangle$ to the density is given through the expectation value (2.32). This establishes the direct potential-density map $\{V_{ext}\} \rightarrow \{n\}$.

TDDFT, in the contrast, deals with the "inverse" map, $\{n\} \rightarrow \{V_{ext}\}$. The questions in TDDFT can be categorized in 3 main categories. First, v-representability problem, does the inverse map exist? Second, uniqueness problem, is the inverse map unique? Third, functional problem, what is the explicit inverse map?

The Runge-Gross theorem, in 1984, [6] answers the second question for a particular case of t-analytic densities. It states that the one-particle density $n(\mathbf{r}, t)$ of a many interacting electron system, for a given initial state, is indeed uniquely determined by the driving potential, provided the density is v-representable .

---

[1]A derivation on the continuity equation in a lattice is given in the appendix A.1

Theorem 2.2.1 (Runge-Gross). For every single particle potential $v(\mathbf{r})$ which can be expanded into a Taylor series with respect to the time around $t = t_0$ a map $G : v(\mathbf{r}, t) \to n(\mathbf{r}, t)$ is defined by solving the time-dependent Schrödinger equation with a fixed initial state $|\Psi_0\rangle$ and calculating the corresponding densities $n(\mathbf{r}, t)$. This map can be inverted up to an additive merely time-dependent function.

Proof. Let us assume two identical quantum systems evolving from the same initial state $|\Psi_0\rangle$ and thus having the same initial density $n(\mathbf{r}, t_0)$. We prove that at $t > 0$ these two systems have different time-dependent densities $n(\mathbf{r}, t) \neq n'(\mathbf{r}, t)$ if they are driven by two Taylor expandable external potentials, $v(\mathbf{r}, t)$ and $v'(\mathbf{r}, t)$, which differ by more than a time-dependent constant, see Fig. 2.1

$$v(\mathbf{r}, t) - v'(\mathbf{r}, t) \neq c(t). \tag{2.37}$$
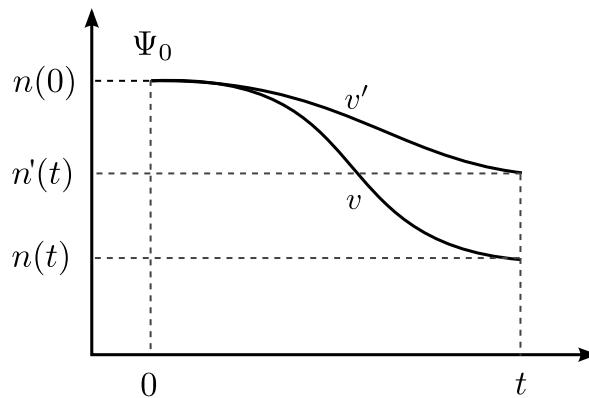
FIGURE 2.1: Schematic graph of the TDDFT mapping stated in the Runge-Gross Theorem 2.2.1.

Therefore there is a smallest integer k such that the k-th time derivative differs by more than a constant

$$\frac{\partial^k}{\partial t^k}[v(\mathbf{r}, t) - v'(\mathbf{r}, t)]|_{t=0} \neq C. \tag{2.38}$$

Next, we show that consequently the $(k + 2)$-th time derivative of the densities, $n(\mathbf{r}, t)$ and $n'(\mathbf{r}, t)$, are different.

The first step is to prove that these two different external potentials, v and v', lead to different time-dependent currents. So, using the Heisenberg equation (2.33), we write

the equations of motion for the currents driven by the potentials

$$\frac{\partial}{\partial t}\mathbf{j}(\mathbf{r},t) = -i\langle\Psi(t)|[\hat{\mathbf{j}}(\mathbf{r}),\hat{H}(t)]|\Psi(t)\rangle, \tag{2.39}$$

$$\frac{\partial}{\partial t}\mathbf{j}'(\mathbf{r},t) = -i\langle\Psi'(t)|[\hat{\mathbf{j}}(\mathbf{r}),\hat{H}'(t)]|\Psi'(t)\rangle, \tag{2.40}$$

where $\hat{H}$ and $\hat{H}'$ are the Hamiltonians (2.30) with v and v' respectively.

Then we calculate the difference of the current derivatives at the initial time, $t = 0$:

$$\frac{\partial}{\partial t}\left(\mathbf{j}(\mathbf{r},t) - \mathbf{j}'(\mathbf{r},t)\right)|_{t=0} = -i\langle\Psi_0|[\hat{\mathbf{j}}(\mathbf{r}),\hat{H}(0) - \hat{H}'(0)]|\Psi_0\rangle. \tag{2.41}$$

The kinetic $\hat{T}$ and interaction $\hat{W}$ cancels between the Hamiltonians and the only remaining term is the difference between the external potentials which can be easily calculated

$$\frac{\partial}{\partial t}\left(\mathbf{j}(\mathbf{r},t) - \mathbf{j}'(\mathbf{r},t)\right)|_{t=0} = -n(\mathbf{r},0)\nabla\left(v_{ext}(\mathbf{r},0) - v'_{ext}(\mathbf{r},0)\right). \tag{2.42}$$

Assuming the density $n(\mathbf{r},0)$ is non-zero [2], the condition Eq. (2.38) is fulfilled for $k = 0$. Otherwise we need to go beyond the first derivative and calculate the difference between higher derivatives of the currents until we encounter a non-zero difference. So calculating $(k + 1)$-th derivative of the current difference we have

$$\left(\frac{\partial}{\partial t}\right)^{k+1}\left(\mathbf{j}(\mathbf{r},t) - \mathbf{j}'(\mathbf{r},t)\right)|_{t=0} = -n(\mathbf{r},0)\nabla u_k(\mathbf{r},0), \tag{2.43}$$

where we define a new function u_k

$$u_k(\mathbf{r},0) = \frac{\partial^k}{\partial t^k}\left(v(\mathbf{r},t) - v'(\mathbf{r},t)\right)|_{t=0}. \tag{2.44}$$

By assumption (2.38) there is such k that $u_k(\mathbf{r},0)$ is non-constant. We also assumed the density is nodeless $n(\mathbf{r},t) \neq 0$. Therefore the $(k+1)$-th derivatives are different and this proves the currents, $\mathbf{j}$ and $\mathbf{j}'$, are different infinitesimally after $t = 0$.

By now, we showed that different potentials lead to different currents. However, we need to prove that the difference in the currents always translates to the densities.

---

[2]This is an extra assumption which is not required by the original Runge-Gross proof. Nonetheless this is a common assumption in density functional type theories

By taking divergence of the equation (2.43) and using the continuity equation (2.35) we get the time derivative of the density difference

$$\left(\frac{\partial}{\partial t}\right)^{k+2} \left(n(\mathbf{r}, t) - n'(\mathbf{r}, t)\right)|_{t=0} = \nabla \cdot [n(\mathbf{r}, 0)\nabla u_k(\mathbf{r}, t)]. \tag{2.45}$$

The right hand side of this equation is zero only if the term in the square brackets are purely rotational. To show that this cannot be the case let us calculate the following integral

$$\int d^3\mathbf{r} u_k(\mathbf{r}, 0)\nabla \cdot [n(\mathbf{r}, 0)\nabla u_k(\mathbf{r}, 0)] = -\int d^3\mathbf{r} n(\mathbf{r}, 0) \left[\nabla u_k(\mathbf{r}, 0)\right]^2$$
$$+ \oint d\mathbf{S} \cdot [u_k(\mathbf{r}, 0)n(\mathbf{r}, 0)\nabla u_k(\mathbf{r}, 0)]. \tag{2.46}$$

By sending the surface of the integral to infinity the second integral in the right hand side goes to zero presuming that the density falls fast enough. By assumption, there is some $\mathbf{r}$ that the integrand of the first integral is non-zero and always non-negative. Thus the whole integral is positive. This means there must be some $\mathbf{r}$ where $\nabla \cdot [n(\mathbf{r}, 0)\nabla u_k(\mathbf{r}, 0)] \neq 0$. This proves that n and n' are different just infinitesimally after the initial time $t = 0$. □

Assumption of the Runge-Gross theorem

In the Runge-Gross theorem the density-potential mapping was proven under some essential assumptions. However it seems that the result of this theorem is valid well beyond some of the assumptions. Below we list the assumptions of the Runge-Gross theorem and proof, some more obvious and some more subtle:

- The time-dependent density $n(\mathbf{r}, t)$ is v-representable at $t > 0$.

- The potential $v(\mathbf{r}, t)$ is Taylor expandable around the initial time $t = 0$.

- The time-dependent density $n(\mathbf{r}, t)$ is analytic in t.

- The density is nodeless $n(\mathbf{r}, t) \neq 0$.

- The system is finite, i.e. there is some finite r_0 such that $n(\mathbf{r}, t) = 0$ for $r \geq r_0$.

2.3 Alternative formulation of the problem, Non-linear Schrödinger equation formalism

The Runge-Gross theorem answers the uniqueness question using the Taylor expansion; however, it does not suggest any systematic way to invert the map or, in other words, to find the potential from the density. Here we formulate the density-potential mapping in terms of a set of closed equations where its solution determines the universal density potential map.

The Schrödinger equation (2.29) defines the direct map from the external potential to the wave function and the density. The strategy is to find an additional equation which links the density to the external potential and/or the wave function. Therefore the resulting equation together with the Schrödinger equation can form a closed set of equations for the external potential and the wave function in terms of given density and initial state.

Although the continuity equation (2.34) determines the first derivative of the density for a given wave function, it does not uniquely fix the current for a given density, as a result of the divergence in the right hand side , so we cannot solve it for the wave function in terms of the density.

Differentiating the continuity equation (2.35) with respect to t results in an equation which plays a key role in TDDFT

$$\ddot{n}(\mathbf{r}, t) = \nabla \cdot (n(\mathbf{r}, t)\nabla v(\mathbf{r}, t)) + i\nabla \cdot \langle\Psi(t)|[\hat{\mathbf{j}}(\mathbf{r}, t), \hat{T} + \hat{W}]|\Psi(t)\rangle. \qquad (2.47)$$

This equation is the divergence of the so called force balance equation, however, from now on we follow the standard practice and simply call it the force balance equation. The force balance equation is a hydrodynamical equation which states that the acceleration for a volume element of a liquid is equal to the sum of the external forces and the internal forces and the stress force, in the liquid. The first term in the right hand side of the force balance equation (2.47) is the divergence of the external force, the Sturm-Liouville term, and the second term is the divergence of the stress force. The force balance equation plays a central role in the TDDFT mappings. It connects the density n to the external potential v and the wave function $|\Psi(t)\rangle$. By rearranging the force balance equation we

get an equation which relates the external potential v_{ext} to the density and the wave function $|\Psi(t)\rangle$.

The force balance equation together with the Schrödinger equation (2.29) form a closed set of closed equations for potential v, wave function $|\Psi(t)\rangle$ provided the density n and the initial state $|\Psi_0\rangle$ are known

$$i\partial_t|\Psi(t)\rangle = \hat{H}[v_{ext}]|\Psi(t)\rangle, \tag{2.48}$$

$$\nabla \cdot (n(\mathbf{r},t)\nabla v_{ext}(\mathbf{r},t)) = \ddot{n}(\mathbf{r},t) - i\nabla \cdot \langle\Psi(t)|\hat{\mathbf{j}}(\mathbf{r},t), \hat{T} + \hat{W}]|\Psi(t)\rangle. \tag{2.49}$$

Consequently the v-representability and uniqueness problems are reduced to the existence of a unique solution to this system of equations for a given pair of initial state $|\Psi_0\rangle$ and time-dependent density n.

Importantly the density n and initial state $|\Psi_0\rangle$ are not independent of each other and fixing $|\Psi_0\rangle$ uniquely determines n and $\dot{n}$

$$\langle\psi_0|\hat{n}(\mathbf{r})|\psi_0\rangle = n(\mathbf{r};t_0), \tag{2.50}$$

$$-\nabla\langle\psi_0|\hat{\mathbf{j}}(\mathbf{r})|\psi_0\rangle = \dot{n}(\mathbf{r};t_0). \tag{2.51}$$

This puts a condition on the initial state $|\Psi_0\rangle$ and the density n which, means that densities which are not consistent with the initial state $|\Psi_0\rangle$ are non-v-representable .

There are two ways suggested in the literature to tackle v-representability and uniqueness problem using the system of equations (2.48). First one is known as the fixed point approach. [17] In this approach one assumes a time-dependent density $n(\mathbf{r},t)$ consistent with an initial state $|\Psi_0\rangle$. Then in an iterative procedure, starts with a guess potential v_0 and solves the Schrödinger equation with the initial state $|\Psi_0\rangle$ to find $|\psi^0(t)\rangle$. Then by solving the force balance with $n(\mathbf{r},t)$ and $|\psi^0(t)\rangle$ he/she gets a potential v_1 which will substitute it back into the Schrödinger equation . It is proven in Ref. [17] that this iteration converges to a unique potential v provided that the response function of the divergence of the internal forces is bounded.

In the second approach one solves the force balance (2.49) equation to find the external potential v_{ext} in term of the density n and the wave function $|\Psi(t)\rangle$

$$v_{ext}[n, \Psi] = (\nabla \cdot n(\mathbf{r}, t)\nabla)^{-1} \left[\ddot{n}(\mathbf{r}, t) - i\nabla \cdot \langle \Psi(t)|\hat{\mathbf{j}}(\mathbf{r}, t), \hat{T} + \hat{W}]|\Psi(t)\rangle \right]. \quad (2.52)$$

Substituting it back into the Schrödinger equation results a non-linear Schrödinger equation (NLSE) with a cubic non-linearity in the wave function $|\Psi(t)\rangle$. [9, 104] In this case the v-representability and uniqueness problems are reduced into existence of a unique solution to NLSE for a given initial state $|\Psi_0\rangle$ and density n.

In both approaches one needs to invert the Sturm-Liouville operator, Eq. (2.52), which is in general a non-trivial problem. Fortunately it has been shown recently that for a bounded domain with a periodic or zero boundary condition the Sturm-Liouville operator is guaranteed to be invertible. [112, 113] So the only thing that remains is to study the Lipschitz continuity of $v_{ext}[n, \Psi]$ (2.52) and then using the well established theorems for quasilinear PDE's [114, 115] we will be able to fully answer both the v-representability and uniqueness questions in TDDFT.

However, for the moment, there is an alternative way to approach the problem. By approximating the infinite-dimensional Hilbert space with a finite-dimensional one, e.g. tight-binding approximation, the Sturm-Liouville operator reduces to a matrix with finite dimensions which can be easily "inverted". [3] In addition NLSE reduces to a system of ordinary differential equations (ODE) which is much easier to handle. By using this approach in chapter 3 we prove the basic theorems for the lattice TDDFT. Then in chapter 4 we will extend those theorems into electronic systems interacting with quantized electric field.

2.4 Electronic system interacting with quantized electromagnetic field

By now we introduced the density-potential mapping for the system of interacting particles in a classical electric field. In this construction the electric field is treated as an external variable where its value is determined by a time-dependent scalar function v_{ext}.

---

[3]The invertibility of the discretized Sturm-Liouville operator is discussed in chapter 3.

However, despite its generality, this construction is an approximation to a more accurate setup in which we consider the quantum mechanical nature of electromagnetic field. For example the quantum electrodynamical (QED) description of the light becomes essential when we are studying a Fermionic system in an optical cavity. [29–31, 37–39]

In this section we present a brief introduction to the non-relativistic photonic Hamiltonian. We start with the wave equation in the classical electromagnetism and then by defining canonical variables we quantize the energy of the field. Afterwards using the radiation Hamiltonian we write the full Hamiltonian for a Fermionic system in an optical cavity. The discrete version of the electron-photon Hamiltonian introduced here is later used in chapter 4 where we prove the basic QED-TDDFT theorems on a lattice.

2.4.1 Canonical quantization

The time evolution of the classical electromagnetic field in vacuum is given by the Maxwell equations for free field

$$\nabla \times \mathbf{E} + \partial_t \mathbf{B} = 0, \tag{2.53a}$$

$$\nabla \times \mathbf{B} - \partial_t \mathbf{E} = 0, \tag{2.53b}$$

$$\nabla \cdot \mathbf{E} = 0, \tag{2.53c}$$

$$\nabla \cdot \mathbf{B} = 0. \tag{2.53d}$$

The electric $\mathbf{E}$ and magnetic field $\mathbf{B}$ can be expressed in terms of the scalar v and vector $\mathbf{A}$ potential

$$\mathbf{B} = \nabla \times \mathbf{A}' \tag{2.54a}$$

$$\mathbf{E} = -\partial_t \mathbf{A} + \nabla v, \tag{2.54b}$$

where $\mathbf{E}$ and $\mathbf{B}$ are invariant under the following gauge transformation

$$\mathbf{A} \to \mathbf{A}' + \nabla \chi, \tag{2.55a}$$

$$v \to v' - \partial_t \chi. \tag{2.55b}$$

Therefore it is possible to choose a gauge where the scalar potential is zero everywhere $v = 0$, the temporal gauge.

Using the Maxwell equations (2.53) one can show that the vector potential obeys the wave equation

$$\nabla^2 \mathbf{A} - \partial_t^2 \mathbf{A} = 0. \tag{2.56}$$

Considering the periodic boundary condition we can easily write the vector potential in terms of plane waves

$$\mathbf{A}(\mathbf{r}, t) = \sqrt{\frac{2\pi}{L^3}} \sum_{\mathbf{k}, \lambda} \frac{\boldsymbol{\varepsilon}_{\mathbf{k}\lambda}}{k} \left[e^{i\beta} e^{i\mathbf{k}\cdot\mathbf{r}} a_{\mathbf{k}\lambda}(t) + e^{-i\beta} e^{-i\mathbf{k}\cdot\mathbf{r}} a_{\mathbf{k}\lambda}^*(t) \right], \tag{2.57}$$

where L^3 is the volume of the cavity, $\boldsymbol{\varepsilon}_{\mathbf{k}\lambda}$ is the polarization vector for the mode with wave vector $\mathbf{k}$ and polarization λ, and β is an arbitrary constant phase. The time-dependent coefficient $a_{\mathbf{k}\lambda}(t)$ for each mode is given by

$$a_{\mathbf{k}\lambda}(t) = e^{i\omega_{\mathbf{k}} t} a_{\mathbf{k}\lambda}, \tag{2.58}$$

where the constant $a_{\mathbf{k}\lambda}$ is fixed through the initial value and boundary conditions. The vector potential $\mathbf{A}$ of (2.57) fulfills the wave equation (2.56) if $\omega_k = k$ and $\mathbf{k} \cdot \boldsymbol{\varepsilon}_{\mathbf{k}\lambda} = 0$.

Substituting $\mathbf{A}(\mathbf{r}, t)$ of (2.57) back into the equations for the electric $\mathbf{E}$ and magnetic field $\mathbf{B}$ (2.59) we get

$$\mathbf{E}(\mathbf{r}, t) = \sqrt{\frac{2\pi}{L^3}} \sum_{\mathbf{k}, \lambda} \frac{\boldsymbol{\varepsilon}_{\mathbf{k}\lambda}}{k} \left[e^{i(\beta + \pi/2)} e^{i\mathbf{k}\cdot\mathbf{r}} a_{\mathbf{k}\lambda}(t) + c.c. \right], \tag{2.59a}$$

$$\mathbf{B}(\mathbf{r}, t) = \sqrt{\frac{2\pi}{L^3}} \sum_{\mathbf{k}, \lambda} \frac{\boldsymbol{\varepsilon}_{\mathbf{k}\lambda} \times \mathbf{k}}{k} \left[e^{i(\beta + \pi/2)} e^{i\mathbf{k}\cdot\mathbf{r}} a_{\mathbf{k}\lambda}(t) + c.c. \right]. \tag{2.59b}$$

Now we can write the Hamiltonian of the classical radiation field. The energy of the radiation field is given by

$$U = \frac{1}{8\pi} \int \left(E^2 + B^2 \right) d\mathbf{r}. \tag{2.60}$$

Therefore substituting the expressions of the Electric and magnetic fields (2.59) we get

$$U = \sum_{\mathbf{k},\lambda} \omega_k a_{\mathbf{k}\lambda}^* a_{\mathbf{k}\lambda} = \frac{1}{2} \sum_{\mathbf{k},\lambda} \omega_k \left(a_{\mathbf{k}\lambda}^* a_{\mathbf{k}\lambda} + a_{\mathbf{k}\lambda} a_{\mathbf{k}\lambda}^*\right). \tag{2.61}$$

Of course the constants $a_{\mathbf{k}\lambda}^*$ and $a_{\mathbf{k}\lambda}$ in the right hand side commute here but to quantize the field we need to write everything in a symmetric manner. It also makes sense to write the energy U in terms of canonical variables before quantization

$$q_{\mathbf{k}\lambda} = \left(\frac{1}{2\omega_{\mathbf{k}}}\right)^{\frac{1}{2}} \left(a_{\mathbf{k}\lambda} + a_{\mathbf{k}\lambda}^*\right), \tag{2.62}$$

$$p_{\mathbf{k}\lambda} = -i\left(\frac{\omega_{\mathbf{k}}}{2}\right)^{\frac{1}{2}} \left(a_{\mathbf{k}\lambda} - a_{\mathbf{k}\lambda}^*\right). \tag{2.63}$$

The field energy in terms of the new canonical coordinates become

$$U = \frac{1}{2} \sum_{\mathbf{k},\lambda} \left(p_{\mathbf{k}\lambda}^2 + \omega_{\mathbf{k}}^2 q_{\mathbf{k}\lambda}^2\right). \tag{2.64}$$

$p_{\mathbf{k}\lambda}$ and $q_{\mathbf{k}\lambda}$ are called canonical variables since they follow

$$\dot{q}_{\mathbf{k}\lambda} = \frac{\partial U}{\partial p_{\mathbf{k}\lambda}}, \quad \dot{p}_{\mathbf{k}\lambda} = -\frac{\partial U}{\partial q_{\mathbf{k}\lambda}}. \tag{2.65}$$

To quantize the field we substitute the canonical variables with non-commutative operators

$$[\hat{q}_{\mathbf{k}\lambda}, \hat{p}_{\mathbf{k}'\lambda'}] = i\delta_{\mathbf{k}\mathbf{k}'}\delta_{\lambda\lambda'} \tag{2.66}$$

Consequently the field energy U is promoted to the Hamiltonian for the field

$$\hat{H}_f = \frac{1}{2} \sum_{\mathbf{k},\lambda} \left(\hat{p}_{\mathbf{k}\lambda}^2 + \omega_{\mathbf{k}}^2 \hat{q}_{\mathbf{k}\lambda}^2\right). \tag{2.67}$$

The Hamiltonian for the free field is nothing but the Hamiltonian of an ensemble of harmonic oscillators with frequencies $\omega_{\mathbf{k}}$, where each $\omega_{\mathbf{k}}$ is twofold degenerate with respect to the polarization λ.

Converting $a_{\mathbf{k}\lambda}$ and $a_{\mathbf{k}\lambda}^*$ to non-commuting operators in the field energy U (2.49) results the field Hamiltonian in the second quantization

$$\hat{H}_f = \frac{1}{2} \sum_{\mathbf{k},\lambda} \omega_k \left(\hat{b}_{\mathbf{k}\lambda}^\dagger \hat{b}_{\mathbf{k}\lambda} + \hat{b}_{\mathbf{k}\lambda} \hat{b}_{\mathbf{k}\lambda}^\dagger \right) = \sum_{\mathbf{k},\lambda} \omega_k \left(\hat{b}_{\mathbf{k}\lambda}^\dagger \hat{b}_{\mathbf{k}\lambda} + \frac{1}{2} \right) \tag{2.68}$$

where $\hat{b}_{\mathbf{k}\lambda}^\dagger$ ($\hat{b}_{\mathbf{k}\lambda}$) creates (annihilates) a photon with the wave vector $\mathbf{k}$ and with the polarization λ. The photon creation and annihilation operators fulfill the following commutation relation

$$[\hat{b}_{\mathbf{k}\lambda}, \hat{b}_{\mathbf{k}'\lambda'}^\dagger] = \delta_{\mathbf{k}\mathbf{k}'}\delta_{\lambda\lambda'}. \tag{2.69}$$

Having the field energy quantized in the next section we will write the Hamiltonian for the electromagnetic field interacting with matter.

2.5 N electrons coupled to the quantized light

Let us turn to a problem where a system of N electrons interacts with quantized light. There is an external time-dependent current $\mathbf{J}_{ext}(t)$ which acts as the external perturbation for the field. The following expression describes the Hamiltonian of this system in the temporal gauge (2.54)

$$\hat{H} = \frac{1}{2m} \sum_{j=1}^{N} \left[i\nabla_j + \hat{\mathbf{A}}(\mathbf{r}) \right]^2 + \hat{W} + \hat{H}_f - \mathbf{J}_{ext}(t) \cdot \hat{\mathbf{A}}(\mathbf{r}). \tag{2.70}$$

Where the vector potential operator $\hat{\mathbf{A}}(\mathbf{r})$ is found by replacing the complex functions $a_{\mathbf{k}\lambda}$ and $a_{\mathbf{k}\lambda}^*$ in Eq. (2.57) by the annihilation $\hat{b}_{\mathbf{k}\lambda}$ and creation operator $\hat{b}_{\mathbf{k}\lambda}^\dagger$ respectively. Therefore $\hat{\mathbf{A}}(\mathbf{r})$ in the Schrödinger representation reads as:

$$\hat{\mathbf{A}}(\mathbf{r}) = \sqrt{\frac{2\pi}{L^3}} \sum_{\mathbf{k},\lambda} \frac{\varepsilon_{\mathbf{k}\lambda}}{k} \left[e^{i\mathbf{k}\cdot\mathbf{r}} \hat{b}_{\mathbf{k}\lambda}^\dagger + e^{-i\mathbf{k}\cdot\mathbf{r}} \hat{b}_{\mathbf{k}\lambda}^\dagger \right]. \tag{2.71}$$

In the long wave length limit we can adopt the dipole approximation where $\exp i\mathbf{k}.\mathbf{r} \approx 1$ and so we have for the vector potential

$$\hat{\mathbf{A}} = \sqrt{\frac{2\pi}{L^3}} \sum_{\mathbf{k},\lambda} \frac{\boldsymbol{\varepsilon}_{\mathbf{k}\lambda}}{k} \left[\hat{b}^\dagger_{\mathbf{k}\lambda} + \hat{b}^\dagger_{\mathbf{k}\lambda} \right], \tag{2.72}$$

Using definition for the canonical coordinate $\hat{q}$ (2.62) we can rewrite the vector potential as

$$\hat{\mathbf{A}} = \sqrt{\frac{4\pi}{L^3}} \sum_{\mathbf{k},\lambda} \hat{q}_{\mathbf{k}\lambda} \boldsymbol{\varepsilon}_{\mathbf{k}\lambda}. \tag{2.73}$$

Substituting back $\hat{\mathbf{A}}$ into Eq. (2.70) we get the full Hamiltonian for the system in the temporal gauge

$$\begin{aligned}
\hat{H} = \quad & \frac{1}{2m} \sum_{j=1}^N \left[i\nabla_j + \sum_\alpha \hat{q}_\alpha \boldsymbol{\lambda}_\alpha \right]^2 + \frac{1}{2} \sum_{i \neq j} w(\mathbf{r}_i, \mathbf{r}_j) \\
& + \sum_\alpha \left(\frac{1}{2}\hat{p}_\alpha^2 + \frac{1}{2}\omega_\alpha^2 \hat{q}_\alpha^2 - \mathbf{J}_{ext} \cdot \boldsymbol{\lambda}_\alpha \hat{q}_\alpha \right).
\end{aligned} \tag{2.74}$$

where the α summation is over $\{(\mathbf{k}, \lambda)\}$ and the vector $\boldsymbol{\lambda}$ is a renormalized polarization vector $\boldsymbol{\lambda} = \sqrt{\frac{4\pi}{L^3}} \boldsymbol{\varepsilon}$.

As a result of the dipole approximation the Hamiltonian $\hat{H}$ in the length gauge is fully independent of the vector potential. Therefore doing a simple gauge transformation (2.55) the Hamiltonian transforms to:

$$\begin{aligned}
\hat{H} = \quad & \sum_{j=1}^N \left(\frac{\nabla_j^2}{2m} + \sum_\alpha \boldsymbol{\lambda}_\alpha \cdot \mathbf{r}_j \hat{p}_\alpha \right) + \frac{1}{2} \sum_{i \neq j} w(\mathbf{r}_i, \mathbf{r}_j) \\
& + \sum_\alpha \left(\frac{1}{2}\hat{p}_\alpha^2 + \frac{1}{2}\omega_\alpha^2 \hat{q}_\alpha^2 + \boldsymbol{\lambda}_\alpha \cdot \mathbf{d}_{ext}(t)\hat{p}_\alpha \right),
\end{aligned} \tag{2.75}$$

where $\mathbf{d}_{ext}(t)$ is the dipole moment of the charge accumulated from the external current. $\sum \boldsymbol{\lambda}\hat{p}$ in the first and the last term is the operator form of the electric field $\hat{E}$.

Later in chapter 4 we use the discretized version of Eq. (2.75) in the context of density-potential mapping for systems interacting with quant light.

Chapter 3

Time-dependent density functional theory on a lattice

3.1 Introduction

In the previous chapter we showed that the v-representability and uniqueness problems in TDDFT are equivalent to the existence of a unique solution to the universal NLSE which determines the potential and the many-body wave function in terms of a given basic observable like density. Mathematically this universal problem can be formulated in two distinct ways. One possibility is to formulate it as a fixed point problem. [17, 18] However here we pose it alternatively as a Cauchy problem for the NLSE. [9, 13–16] We apply the NLSE technique of Ref. [16] for the lattice TDCDFT to address the TDDFT mapping problem on a lattice.

First we present a proof of uniqueness and existence theorems for the lattice TDDFT and analyze conditions which have to be imposed on the initial state and the density to guarantee v-representability . Afterwards a special case will be discussed where we show any properly normalized density is locally v-representable provided the dynamics starts from the ground state. Then we clarify the problem further by an example of a single particle on a two-site lattice.

This chapter is structured as the following. In section 3.2 we present a general formulation of the lattice many-body theory and derive a lattice analog of the force balance equation that plays a key role in the NLSE formalism for TDDFT. section 3.3 is the central part of this chapter. Subsection 3.3.1 starts with a brief overview of our approach to TDDFT in a more familiar continuum case. Then we derive the corresponding NLSE for a lattice theory, and finally, formulate and prove the general existence of a uniqueness theorem on the lattice TDDFT. Several important aspects of the basic theorem for a generic initial state are discussed in subsection 3.3.2. Section 3.4 presents an explicit illustration of the general existence theorem for a simple exactly solvable model – one particle on a two-site lattice. In section 3.5 we consider a practically important case of a system evolving from its ground state. The main outcome of this section is a v-representability theorem for the initial ground state.

3.2   Preliminaries: Many-body problem on a lattice

Let us consider dynamics of a system of N interacting quantum particles on a lattice that consists of a finite but arbitrary large number M of sites. The many-body wave function $\psi(\mathbf{r}, \mathbf{r}_2, ..., \mathbf{r}_N; t)$ characterizes the state of the system at time t , where coordinates $\mathbf{r}_i$ of particles $(i = 1, 2, .., N)$ take values on the lattice sites. And the following Schrödinger equation describes dynamics driven by an external on-site potential $v(\mathbf{r}; t)$:

$$
\begin{aligned}
i\partial_t \psi(\mathbf{r}_1, ..., \mathbf{r}_N; t) = \ & - \sum_{i=1}^{N} \sum_{\mathbf{x}_i} T_{\mathbf{r}_i, \mathbf{x}_i} \psi(..., \mathbf{x}_i, ...; t) \\
& + \sum_{j=1}^{N} v(\mathbf{r}_j; t) \psi(\mathbf{r}_1, ..., \mathbf{r}_N; t) \\
& + \sum_{j>i} w_{\mathbf{r}_i, \mathbf{r}_j} \psi(\mathbf{r}_1, ..., \mathbf{r}_N; t), \quad\quad (3.1)
\end{aligned}
$$

where real coefficients $T_{\mathbf{r}, \mathbf{r}'} = T_{\mathbf{r}', \mathbf{r}}$ correspond to the rate of hopping from site $\mathbf{r}$ to site $\mathbf{r}'$ (we assume $T_{\mathbf{r}, \mathbf{r}} = 0$), and $w_{\mathbf{r}, \mathbf{r}'}$ is a potential of a pairwise particle-particle interaction. Here we do not specify the geometry of the lattice and neither assume that the interaction depends only on the distance between particles. For example, the latter

is important in a typical transport setup where non-interacting or weakly interacting leads connected to a strongly interacting central region . [1][25, 27, 28, 116, 117]

Equation (3.1) determines the wave function as a unique functional of continuous and bounded external potential and a given initial state,

$$\psi(\mathbf{r}_1, \mathbf{r}_2, ..., \mathbf{r}_N; t_0) = \psi_0(\mathbf{r}_1, \mathbf{r}_2, ..., \mathbf{r}_N). \tag{3.2}$$

As we discussed in the previous chapter the density $n(\mathbf{r}; t)$ of particles is the basic variable in TDDFT. However in the present context it corresponds to the number of particles on a given site and it is defined as:

$$n(\mathbf{r}; t) = N \sum_{\mathbf{r}_2, ..., \mathbf{r}_N} |\psi(\mathbf{r}, \mathbf{r}_2, ..., \mathbf{r}_N; t)|^2, \tag{3.3}$$

where we assumed that the particles are identical. By taking the time derivative of the density's definition (3.3) and using the Schrödinger equation (3.1), we find the following equation of motion for the density

$$\dot{n}(\mathbf{r}; t) = i \sum_{\mathbf{r}'} [T_{\mathbf{r}, \mathbf{r}'} \rho(\mathbf{r}, \mathbf{r}'; t) - T_{\mathbf{r}', \mathbf{r}} \rho(\mathbf{r}', \mathbf{r}; t)], \tag{3.4}$$

where '$\dot{n} = \partial_t n$, and $\rho(\mathbf{r}, \mathbf{r}'; t)$ is a density matrix on the $[\mathbf{r}, \mathbf{r}']$-link,

$$\rho(\mathbf{r}, \mathbf{r}'; t) = N \sum_{\mathbf{r}_2, ..., \mathbf{r}_N} \psi^*(\mathbf{r}', \mathbf{r}_2, ..., \mathbf{r}_N; t) \psi(\mathbf{r}, \mathbf{r}_2, ..., \mathbf{r}_N; t). \tag{3.5}$$

A derivation for the continuity equation on a lattice can be found in the appendix A.1

Equation (3.4) is the lattice version of the continuity equation (2.35). Since in the left hand side of Eq. (3.4) we have the time derivative of the on-site number of particles, the right hand side should be identified with the sum of outgoing currents flowing along links attached to the site. Indeed introducing a link current from site $\mathbf{r}$ to site $\mathbf{r}'$ as follows

$$J(\mathbf{r}, \mathbf{r}') = 2\mathrm{Im}[T_{\mathbf{r}, \mathbf{r}'} \, \rho(\mathbf{r}, \mathbf{r}'; t)], \tag{3.6}$$

---

[1] Formally Eq. (3.1) corresponds to a system of M^N linear ordinary differential equations (ODE). For example, in the case of a single particle on a two-site lattice it is a system of two coupled equations.
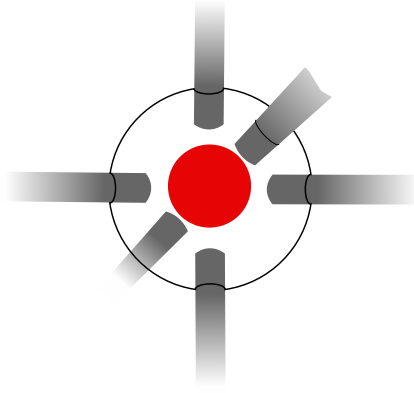
FIGURE 3.1: The continuity equation (3.4) can be understood as an integral over the continuum continuity equation integrated over a given volume surrounding the site i. Therefore the term $\nabla \cdot \mathbf{J}$ would transform to a surface integral over the boundary of the volume which results sum of outgoing integral from the surface.

we can rewrite the continuity equation (3.4) in a more familiar way

$$\dot{n}(\mathbf{r};t) = -\sum_{\mathbf{r}'} J(\mathbf{r},\mathbf{r}'). \tag{3.7}$$

This equation shows that the rate which the density decreases on each site is equal to the sum of all outgoing currents. Equation (3.7) can be also viewed as an integral of the usual differential continuity equation (2.35) over a small volume element surrounding the site $\mathbf{r}$. See Fig. 3.1.

Now we need to introduce another equation of primary importance for the lattice TDDFT. This is a lattice analogue of a divergence of the local force balance equation (2.47). Like before It can be derived by differentiating the continuity equation (3.4) with respect to time, and using Eq. (3.1) to transform the derivative of the right hand side. After straightforward calculations, the lattice force balance equation reduces to the following form

$$\ddot{n}(\mathbf{r};t) = 2\mathrm{Re}\sum_{\mathbf{r}'} T_{\mathbf{r},\mathbf{r}'}\rho(\mathbf{r},\mathbf{r}';t)[v(\mathbf{r}';t) - v(\mathbf{r};t)] + q(\mathbf{r};t). \tag{3.8}$$

Here $q(\mathbf{r};t)$ stands for a "lattice divergence" of the internal stress force,

$$\begin{aligned} q(\mathbf{r};t) \;=\; & -2\mathrm{Re}\sum_{\mathbf{r}',\mathbf{r}''} T_{\mathbf{r},\mathbf{r}'}\Big\{\rho_2(\mathbf{r},\mathbf{r}'',\mathbf{r}';t)(w_{\mathbf{r},\mathbf{r}''} - w_{\mathbf{r}',\mathbf{r}''}) \\ & + \; [T_{\mathbf{r}',\mathbf{r}''}\rho(\mathbf{r},\mathbf{r}'';t) - T_{\mathbf{r},\mathbf{r}''}\rho(\mathbf{r}',\mathbf{r}'';t)]\Big\}, \end{aligned} \tag{3.9}$$

where $\rho_2(\mathbf{r}, \mathbf{r}'', \mathbf{r}'; t)$ in the right hand side is the two body density matrix,

$$
\begin{aligned}
\rho_2(\mathbf{r}, \mathbf{r}'', \mathbf{r}'; t) = & \ N \quad (N-1) \sum_{\mathbf{r}_3, \ldots, \mathbf{r}_N} \psi^*(\mathbf{r}, \mathbf{r}'', \ldots, \mathbf{r}_N; t) \\
& \times \quad \psi(\mathbf{r}', \mathbf{r}'', \ldots, \mathbf{r}_N; t).
\end{aligned} \tag{3.10}
$$

A detailed derivation of the continuity equation can be found in the appendix A.2.

A special role of the force balance equation (3.8) for TDDFT follows from the fact that it explicitly relates the potential $v(\mathbf{r}; t)$ to the density $n(\mathbf{r}; t)$ and the instantaneous many-body state $\psi(t)$. As we already discussed in the previous chapter it has a defining role in the TDDFT and together with the Schrödinger equation form a non-linear Schrödinger equation . This equation (3.8) is the main result of the present section, which we will use in the next section to analyze the existence of the lattice TDDFT.

3.3 TDDFT on a lattice

The whole concept of TDDFT is based on the existence of a 1:1 map between the time dependent density and the external potential. In this section we will present the mapping and the v-representability theorems for the lattice TDDFT which were published in Ref. [106].

3.3.1 Statement of the problem and the basic existence theorem

Here we will use the NLSE approach to TDDFT-type theories [9, 13, 15, 16] introduced in the chapter II.

In this framework the proof of the existence of TDDFT reduces to proving the uniqueness and existence of solutions to a nonlinear many-body problem, e.g., NLSE in Eqs. (2.48) in real space supplemented with an initial condition which satisfies the consistency relations of Eqs. (2.50) and (2.51).

Strategically the solution of the outlined nonlinear problem contains two major steps: (i) inverting the Sturm-Liouville operator in the left hand side of Eq. (2.49) to find the potential as a functional of a given density and the instantaneous state, $v[n(t), |\psi(t)\rangle](\mathbf{r})$; and (ii) inserting this potential into Eq. (2.48) and solving the resulting NLSE. In a

continuum proving the corresponding existence theorems for either step is a highly non-trivial and currently unsolved problem, although a significant progress has been made recently [17, 18, 118]. However the problems (i)-(ii) for the lattice many-body theory due to the finite dimensionality of the Hilbert space is more manageable and therefore a rigorous proof of existence and uniqueness can be given.

On a lattice the construction of the inverse map $\{n(t), \Psi_0\} \mapsto \{v(t), \Psi(t)\}$ consists of solving the discrete Schrödinger equation (3.1), where the potential $v(\mathbf{r}; t)$ is determined self-consistently from the lattice force balance equation of Eq. (3.8) which we rewrite as follows

$$\sum_{\mathbf{r}'} k_{\mathbf{r},\mathbf{r}'}[\psi(t)]v(\mathbf{r};t) = \ddot{n}(\mathbf{r};t) - q[\psi(t)](\mathbf{r}). \tag{3.11}$$

Here the functional $q[\psi(t)](\mathbf{r})$ is defined by Eq. (3.9) and we introduced the following notation

$$k_{\mathbf{r},\mathbf{r}'}(\psi) = 2\mathrm{Re}\left[T_{\mathbf{r},\mathbf{r}'}\rho(\mathbf{r},\mathbf{r}') - \delta_{\mathbf{r},\mathbf{r}'}\sum_{\mathbf{r}''}T_{\mathbf{r},\mathbf{r}''}\rho(\mathbf{r},\mathbf{r}'')\right]. \tag{3.12}$$

The initial condition for the nonlinear problem of Eq. (3.1) and (3.11) should satisfy the consistency conditions which follow from the definition of the density Eq. (3.3) and the continuity equation (3.2)

$$N\sum_{\mathbf{r}_2,...,\mathbf{r}_N} |\psi_0(\mathbf{r},\mathbf{r}_2,...,\mathbf{r}_N)|^2 = n(\mathbf{r};t_0), \tag{3.13}$$

$$-2\mathrm{Im}\sum_{\mathbf{r}'}T_{\mathbf{r},\mathbf{r}'}\rho_0(\mathbf{r},\mathbf{r}') = \dot{n}(\mathbf{r};t_0). \tag{3.14}$$

Equations (3.1), (3.11), (3.13), and (3.14) are the lattice analogs of Eqs. (2.29), (2.47) and (2.50). A dramatic simplification of the lattice theory comes from the fact that both the Hilbert space $\mathcal{H}$ and the space $\mathcal{V}$ of lattice-valued potentials become finite dimensional with dimensions M^N and M, respectively. In particular, the lattice N-body Schrödinger equation (3.1) corresponds to a system of M^N ODE, while the force balance equation (3.11) reduces to a system of M algebraic equations. In fact, Eq. (3.11) can be conveniently rewritten in a matrix form

$$\hat{K}(\psi)V = S(\ddot{n},\psi), \tag{3.15}$$

where $\hat{K}$ is a real symmetric $M \times M$ matrix with elements $k_{\mathbf{r},\mathbf{r}'}$ of Eq. (4.33), and V and S are M-dimensional vectors with components

$$v_{\mathbf{r}} = v(\mathbf{r}) \quad \text{and} \quad s_{\mathbf{r}}(\ddot{n}, \psi) = \ddot{n}(\mathbf{r}) - q[\psi](\mathbf{r}), \tag{3.16}$$

respectively. The $\hat{K}$-matrix in Eq. (3.15) is a lattice analog of the Sturm-Liouville operator $m^{-1}\nabla n \nabla$ in Eq. (2.47). Hence on a lattice the step (i) in solving our nonlinear many-body problem reduces to the simple matrix inversion which can be performed if the matrix $\hat{K}$ is nondegenerate. At this point it is worth noting that because of the gauge invariance $\hat{K}$ matrix of Eq. (4.33) it always has at least one zero eigenvalue that corresponds to a space-constant eigenvector. Therefore if $\mathcal{V}$ is the M-dimensional space of lattice potentials $v(\mathbf{r})$, then the invertibility/nondegeneracy of $\hat{K}$ should always refer to the invertibility in the $M-1$-dimensional subspace of $\mathcal{V}$ orthogonal to a constant vector $v_C(\mathbf{r}) = C$. Physically this means that the force balance equation (3.11) determines the self-consistent potential $v[n, \psi](\mathbf{r})$ only up to an arbitrary constant.

Now we are in a position to formulate and prove the basic existence and uniqueness theorem on the lattice TDDFT. All statements of Theorem below refer to the lattice N-body problem defined in section 3.2.

Theorem 3.3.1 (existence of the lattice TDDFT). Assume that a given time-dependent density $n(\mathbf{r}; t)$ is nonnegative on each lattice site, sums up to the number of particles N, and has a continuous second time derivative $\ddot{n}(\mathbf{r}; t)$. Let Ω be a subset of the N-particle Hilbert space $\mathcal{H}$ where the matrix $\hat{K}(\psi)$ of Eq. (4.33) has only one zero eigenvalue corresponding to a space-constant vector. If the initial state $\psi_0 \in \Omega$, and the consistency conditions of Eqs. (3.13) and (3.14) at time t_0 are fulfilled, then

(i) There is a time interval around t_0 where the nonlinear many-body problem of Eqs. (3.1), (3.11) has a unique solution that defines the wave function $\psi(t)$ and the potential $v(t)$ as unique functionals of the density $n(t)$ and initial state ψ_0;

(ii) The solution of item (i) is not global in time if and only if at some maximal existence time $t^* > t_0$ the boundary of Ω is reached.

Proof. By the condition of theorem 3.3.1 ψ_0 belongs to Ω where $\hat{K}(\psi)$ has only one trivial zero eigenvalue. Hence there is a neighborhood of ψ_0, such that for all ψ's from this neighborhood the matrix $\hat{K}(\psi)$ can be inverted (in the $M-1$-dimensional subspace

of $\mathcal{V}$, orthogonal to a constant). In other words, we can solve the force balance equation (3.15) as $V = \hat{K}^{-1}S$ and express (up to a constant) the on-site potential in terms of the instantaneous wave function and the density

$$v[n, \psi](\mathbf{r}) = \sum_{\mathbf{r'}} \hat{K}^{-1}_{\mathbf{r},\mathbf{r'}}(\psi)s_{\mathbf{r}}(\ddot{n}, \psi). \tag{3.17}$$

Substituting this potential into Eq. (3.1) we obtain the following NLSE,

$$
\begin{aligned}
i\partial_t \psi(\mathbf{r}_1, ..., \mathbf{r}_N; t) &= \sum_{j=1}^{N}\sum_{\mathbf{r}} \hat{K}^{-1}_{\mathbf{r}_j,\mathbf{r}}s_{\mathbf{r}}\psi(\mathbf{r}_1, ..., \mathbf{r}_N; t) \\
&- \sum_{i=1}^{N}\sum_{\mathbf{x}_i} T_{\mathbf{r}_i,\mathbf{x}_i}\psi(..., \mathbf{x}_i, ...; t) \\
&+ \sum_{j>i} w_{\mathbf{r}_i,\mathbf{r}_j}\psi(\mathbf{r}_1, ..., \mathbf{r}_N; t).
\end{aligned}
\tag{3.18}
$$

Equation (4.15) supplemented with the initial condition of Eq. (3.2) constitutes a universal nonlinear many-body problem which determines the wave function in terms of the density. Formally it corresponds to a Cauchy problem for a system of $N_{\mathcal{H}} = M^N$ ODE of the following structural form

$$\dot{\psi} = F(\psi, t), \qquad \psi(t_0) = \psi_0, \tag{3.19}$$

where ψ is an $N_{\mathcal{H}}$-dimensional vector living in the Hilbert space $\mathcal{H}$ and the right hand side is a nonlinear function of ψ's components.

The nonlinearity of $F(\psi, t)$ in Eq. (4.15) comes from the dependence of the potential $v[n, \psi](\mathbf{r})$ in Eq. (4.12) on the wave function, which, in turn, is determined by the functions $\hat{K}^{-1}(\psi)$ and $S(\ddot{n}, \psi)$. Both $k_{\mathbf{r},\mathbf{r'}}(\psi)$ of Eq. (4.33), and $s_{\mathbf{r}}(\ddot{n}, \psi)$ of Eq. (4.34) are linear in the density matrices, and thus bilinear in ψ, forms. Therefore the potential $v[n, \psi](\mathbf{r})$ and, as a consequence, the whole right hand side $F(\psi, t)$ in Eq. (3.19) are rational functions of components of the wave function. Moreover, the denominator of these rational functions never turns into zero for all $\psi \in \Omega$, which implies that in Ω the function $F(\psi, t)$ satisfies a uniform Lipshitz condition. An explicit time dependence of $F(\psi, t)$ is determined by the time dependence of $\ddot{n}(t)$ that is continuous by the condition of the theorem. Thus we conclude that for all $\psi \in \Omega$ the right hand side $F(\psi, t)$ in Eq. (3.19) is Lipshitz in ψ and continuous in time.

After identifying the subset Ω with the domain of Lipshitz continuity we can directly employ the standard results of the theory of nonlinear ODE. Namely, if the initial state $\psi_0 \in \Omega$, the Picard-Lindelöf theorem [119] guarantees the existence of a finite interval $t_0 - \delta < t < t_0 + \delta$, with $\delta > 0$, where the initial value problem (3.19) has a unique solution. This solution defines a unique map $\{n(t), \psi_0\} \mapsto \{v(t), \psi(t)\}$ locally in time, in accordance with the statement (i) of the theorem.

The extension theorems for nonlinear ODE imply that the local solution cannot be extended beyond some maximal existence time $t^* > t_0$ only in two cases: first, at $t \to t^*$ the solution becomes unbounded or, second, at $t \to t^*$ it reaches the boundary of Ω. In our case the solution is guarantied to be normalized and thus bounded. Therefore we are left only with the second possibility, which proves the statement (ii) and completes the proof of the theorem. □

We note that in the special case of t-analytic density Picard-Lindelöf theorem reduces to the classical Cauchy theorem for the first order ODE with an analytic nonlinearity. The Cauchy theorem implies that for a given t-analytic density the NLSE procedure returns the wave function and the potential which are also t-analytic. Similarly t-analyticity is preserved in the direct map – a given t-analytic potential always produces a t-analytic density. Therefore the generation of nonanalytic densities by analytic potentials, discussed in Refs.[8–10] for continuum systems, is not possible on a lattice, which is probably related to the boundness of lattice Hamiltonians.

3.3.2 Discussion and comments on the existence theorem

3.3.2.1 Definition of the v-representability subset Ω

According to Theorem 3.3.1, any sufficiently smooth density $n(\mathbf{r}; t)$ is v-representable, at least locally, if the dynamics starts inside the subset Ω of the Hilbert space. In general to ensure that state ψ belongs to Ω we need to check the invertibility of matrix $\hat{K}(\psi)$. Although this is possible in principle, but become difficult in practice, especially for lattices with a large number of sites. Is it possible to formulate simpler, but possibly more restrictive criteria, which would guarantee the validity of the lattice TDDFT.

One simple necessary condition immediately follows from the form of the matrix elements $k_{\mathbf{r},\mathbf{r}'}$ in Eq. (4.33). Matrix $\hat{K}(\psi)$ is nondegenerate only if a lattice state ψ is connected in a sense that any two sites can be connected by a line composed of links with nonzero values of $T_{\mathbf{r},\mathbf{r}'}\mathrm{Re}\rho(\mathbf{r},\mathbf{r}')$. Indeed, for a disconnected state $\hat{K}(\psi)$ takes a block-diagonal form and new zero eigenvalues appear which correspond to piecewise constant in space eigenvectors. The number of such zero eigenvalues equals to the number of disconnected regions on a lattice. We emphasize that a purely geometric connectivity of the lattice does not automatically guaranty v-representability – two sites connected geometrically by a nonzero hopping matrix element $T_{\mathbf{r},\mathbf{r}'}$ can be disconnected in the above sense if for a state ψ the quantity $\mathrm{Re}\rho(\mathbf{r},\mathbf{r}')$ vanishes. An explicit example of such a disconnected (one-particle) state on a connected 4-site tight-binding cluster has been recently proposed in Ref.[22] to demonstrate a possible non-v-representability in the lattice TDDFT. We have considered an excited state with nodes on two opposite corners of a square formed by four sites (see Fig. 1 in Ref. [22]). The two nodes effectively separate the system into two disconnected parts. Therefore the matrix $\hat{K}$ acquires an extra zero eigenvalue and Theorem 3.3.1 does not apply if the dynamics starts from such a state. In fact, one can show that this particular state belongs to the border of the v-representability subset Ω.

Obviously the connectivity of the lattice state is only a necessary but not a sufficient condition for ψ to be in Ω. The reason is that for a connected state the quantities $T_{\mathbf{r},\mathbf{r}'}\mathrm{Re}\rho(\mathbf{r},\mathbf{r}')$ for different links may have different signs which can be responsible for extra zero eigenvalues of $\hat{K}$. Hence the simplest sufficient condition is the connectivity of the state and the positivity (or negativity) of $T_{\mathbf{r},\mathbf{r}'}\mathrm{Re}\rho(\mathbf{r},\mathbf{r}')$ for all lattice links. In other words, a state $\psi \in \Omega$ if its $\hat{K}$ matrix is primitive and does not have a block-diagonal form. This condition is easy to check in practice, but it appears to be quite restrictive. A less restrictive criterion that in many cases can still be checked easily, is a positive (negative) definiteness of $\hat{K}(\psi)$. In section 3.5 we will show that this is exactly the case for a many-body ground state on a connected lattice. Namely, if ψ_0 is a ground state, then $\hat{K}(\psi_0)$ is negative definite and thus $\psi_0 \in \Omega$, which implies the existence of the lattice TDDFT for a system evolving from its ground state.

3.3.2.2 Boundness of $\dot{n}(\mathbf{r};t)$ in the lattice TDDFT

A specific feature of quantum dynamics on a lattice, which narrows the class of v-representable densities, is the boundness of the time derivative of the density [16, 21, 24, 103]. Since the hopping rate along a given link is fixed to be $T_{\mathbf{r},\mathbf{r}'}$, the link current of Eq. (4.7) cannot exceed a certain maximal value, i. e. $|J(\mathbf{r},\mathbf{r}')| \le |J_{\max}(\mathbf{r},\mathbf{r}')|$, where $|J_{\max}|$ can be estimated [16, 20, 24] using the Cauchy-Schwarz inequality

$$|J_{\max}(\mathbf{r},\mathbf{r}')| = 2|T_{\mathbf{r},\mathbf{r}'}\rho(\mathbf{r},\mathbf{r}')| \le 2|T_{\mathbf{r},\mathbf{r}'}|\sqrt{n(\mathbf{r})n(\mathbf{r}')}. \tag{3.20}$$

A physical density should satisfy the continuity equation (3.7) which imposes a bound on its time derivative,

$$|\dot{n}(\mathbf{r})| \le \sum_{\mathbf{r}'} |J_{\max}(\mathbf{r},\mathbf{r}')|. \tag{3.21}$$

At the first glance Theorem 3.3.1 does not say anything about the boundness of $\dot{n}(\mathbf{r};t)$. Therefore it is instructive to see how the latter can be deduced from the conditions of the theorem. First of all we note that if the solution of the universal NLSE exists, then the continuity equation is necessarily satisfied, which can be true only if our given density does not violate the bound of Eq. (3.21). According to the assumptions of the theorem the density should satisfy the consistency conditions, Eqs. (3.13) and (3.14), and its second time derivative $\ddot{n}(\mathbf{r};t)$ should be continuous in time for all $t > t_0$. By imposing the condition of Eq. (3.14) we explicitly require the boundness of $\dot{n}(\mathbf{r};t_0)$ at the initial time t_0, while the continuity of $\ddot{n}(\mathbf{r};t)$ ensures that the physical bound of Eq. (3.21) cannot be violated immediately. It is also worth noting that the boundness of $\dot{n}(\mathbf{r};t)$ is closely related to the invertibility of $hatK$ matrix or, more precisely, to the connectivity of the instantaneous state $\psi(t)$. Indeed, the link current $J(\mathbf{r},\mathbf{r}')$ of Eq. (4.7) and the off-diagonal element $k_{\mathbf{r},\mathbf{r}'}$ of Eq. (4.33) are, respectively, the imaginary and the real parts of the quantity $2T_{\mathbf{r},\mathbf{r}'}\rho(\mathbf{r},\mathbf{r}';t)$. Therefore for any state ψ and $\mathbf{r} \ne \mathbf{r}'$ the following identity holds true

$$|J(\mathbf{r},\mathbf{r}')|^2 + |k_{\mathbf{r},\mathbf{r}'}|^2 = |J_{\max}(\mathbf{r},\mathbf{r}')|^2. \tag{3.22}$$

Equation (3.22) shows that when the current $J(\mathbf{r},\mathbf{r}')$ reaches the maximal value of Eq. (3.20), $k_{\mathbf{r},\mathbf{r}'}$ turns into zero, which breaks the link between sites $\mathbf{r}$ and $\mathbf{r}'$. Hence

saturation of the bound in Eq. (3.21) implies breaking all links attached to a site $\mathbf{r}$. This site becomes disconnected from the rest of the lattice and the $\hat{K}$ matrix acquires an extra zero eigenvalue, indicating that the state ψ is not anymore in the v-representability subset Ω. Thus the saturation of the bound on $\dot{n}(\mathbf{r})$ at some time $t = t^*$ automatically assumes that at this time the solution hits the boundary of Ω. This behavior is in a clear agreement with the statement (ii) of Theorem 3.3.1.

3.4 Explicit illustration: One particle on a two-site lattice

This section is aimed to illustrating the general NLSE scheme using a simple exactly solvable example – one particle on two sites. In spite of its simplicity this example practically contains all features of the most general N-body case, and thus displays all subtle points of the general formulation in a clear and transparent form. Therefore it is advisable to read this section to get a better feeling of the formalism presented in section 3.3. Another explicit example of NLSE for an interacting two-particle system is given in Appendix B.

Consider a particle living on a two-site lattice. The state of the system is described by the one-particle wave function $\psi_{\mathbf{r}}(t)$, where the coordinate $\mathbf{r}$ takes values 1 or 2 corresponding to the two lattice sites.

The dynamics of the system is described by Eq. (4.1) where the number of the particles $N = 1$ and there is no interaction term in the right hand side. Therefore Eq. (4.1) reduces to the following system of two ODE

$$i\partial_t \psi_1 = v_1 \psi_1 - T\psi_2, \tag{3.23a}$$

$$i\partial_t \psi_2 = v_2 \psi_2 - T\psi_1, \tag{3.23b}$$

where T is the hopping rate, and v_1 and v_2 are the time-dependent external potentials on sites 1 and 2, respectively. The system of Eqs. (3.23) determines the components of the wave functions ψ_1 and ψ_2 as functionals of the external potential and the initial state $\psi_{1,2}(0)$ (for brevity we set $t_0 = 0$).

To find the wave function as a functional of the density we have to construct the proper NLSE, and for this we need an additional equation which relates the potential to the

density and wave function. In the general framework of section 3.3 the force balance equation of Eq. (3.8) [or, equivalently, Eq. (3.15)] serves exactly for this purpose. For two sites $\hat{K}$ is a 2×2 matrix and therefore Eq. (3.15) takes the form

$$\begin{pmatrix} -k_{12} & k_{12} \\ k_{12} & -k_{12} \end{pmatrix} \begin{pmatrix} v_1 \\ v_2 \end{pmatrix} = \begin{pmatrix} \ddot{n}_1 - q_1 \\ \ddot{n}_2 - q_2 \end{pmatrix} \tag{3.24}$$

where $k_{\mathbf{rr'}}$ and $q_{\mathbf{r}}$ can easily be derived from the general definitions of Eqs. (4.33) and (3.9), respectively,

$$k_{12} \;\; = \;\; 2T \operatorname{Re}\rho_{12}, \tag{3.25}$$

$$q_1 = -q_2 \;\; = \;\; 2T^2(|\psi_2|^2 - |\psi_1|^2). \tag{3.26}$$

The link density matrix ρ_{12}, which is in general determined by Eq. (4.6), in the present one-particle case reduces to the product $\rho_{12} = \psi_1^* \psi_2$. The 2×2 matrix in the right hand side of Eq. (3.24) is the $\hat{K}$ matrix extensively discussed in the previous section. Obviously $\hat{K}$ always has a zero eigenvalue corresponding to a space-constant potential $v_1 = v_2 = C$. If there is no other zero eigenvalue we can invert $\hat{K}$ in the space perpendicular to the constant vector $v_C(\mathbf{r}) = C$. Being perpendicular to $v_C(\mathbf{r})$ simply means that on-site potentials sum up to zero, which for two sites implies

$$v_1 = -v_2 = v. \tag{3.27}$$

This equation defines a 1-dimensional subspace of $\mathcal{V}$ where $\hat{K}$, in principle, can be inverted. In the present two-site case the $\hat{K}$ matrix is invertible if $k_{12} \neq 0$. Therefore the v-represenatability subset Ω of the Hilbert space is defined by the following simple condition

$$\operatorname{Re}[\psi_1^* \psi_2] \neq 0. \tag{3.28}$$

For all states satisfying the condition Eq. (3.28) we can invert $\hat{K}$ matrix in Eq. (3.24) and find the potential $v = v_1 = -v_2$ as a functional of the density and wave function,

$$v = -\frac{\ddot{n}_1 - 2T^2(|\psi_2|^2 - |\psi_1|^2)}{4T \operatorname{Re}\rho_{12}}. \tag{3.29}$$

where we substituted the explicit expressions for k_{12} and q_1 from Eqs. (3.25) and (3.26).

The final NLSE is obtained by inserting the potential of Eq. (3.29) into the Schrödinger equation (3.23)

$$i\partial_t \psi_1 = -\frac{\ddot{n}_1 - 2T^2(|\psi_2|^2 - |\psi_1|^2)}{4T\mathrm{Re}[\psi_1^* \psi_2]}\psi_1 - T\psi_2, \tag{3.30a}$$

$$i\partial_t \psi_2 = \frac{\ddot{n}_1 - 2T^2(|\psi_2|^2 - |\psi_1|^2)}{4T\mathrm{Re}[\psi_1^* \psi_2]}\psi_2 - T\psi_1. \tag{3.30b}$$

This system of equations perfectly illustrates the generic structure of NLSE appearing in the TDDFT context. Firstly, as described in section 3.3, the nonlinearity is always a rational function with enumerator and denominator having bilinear forms in the components of the wave function ψ. For all $\psi \in \Omega$ [i. e. for ψ satisfying Eq. (3.28)] the denominator never turns to zero. Secondly, the explicit time dependence enters NLSE only via the second time derivative of the density $\ddot{n}_\mathbf{r}(t)$ that is assumed to be continuous. The two properties above ensure that the right hand side of our NLSE is Lipshitz in ψ and continuous in t. By Picard-Lindelöf theorem this guarantees the existence of a unique solution to Eqs. (3.30) for any initial state $\psi(0)$ from Ω.

However, this is not yet the whole story. Since the density enters the equations only via $\ddot{n}_\mathbf{r}(t)$, our unique solution to NLSE, in general, will reproduce only the second time derivative of the prescribed density correctly. The whole externally given density $n_\mathbf{r}(t)$ is recovered from NLSE if the dynamics starts from a special manifold of the "density-consistent" initial states which are defined by the consistency conditions of Eqs. (3.13) and (3.14).

To proceed further with our example, we represent the wave function in the polar form

$$\psi_1(t) = |\psi_1(t)|e^{i\varphi(t)/2}, \ \psi_2(t) = |\psi_2(t)|e^{-i\varphi(t)/2}, \tag{3.31}$$

and substitute it into the consistency conditions. As a result Eqs. (3.13) and (3.14) simplify as follows, respectively,

$$|\psi_\mathbf{r}(0)| = \sqrt{n_\mathbf{r}(0)}, \quad \mathbf{r} = \{1, 2\}, \tag{3.32}$$

$$\dot{n}_1(0) = 2T\sqrt{n_1(0)n_2(0)}\sin\varphi(0), \tag{3.33}$$

where $n_\mathbf{r}(0)$ is the (prescribed) initial density, and $\dot{n}_\mathbf{r}(0)$ is the initial time derivative. The first condition, Eq. (3.32), uniquely determines the modulus of the allowed initial

states in terms of the initial density. Finding the "density-consistent" initial phases is a bit more tricky as the right hand of Eq. (3.33) is not a single valued function. Equation (3.33) has two solutions which can be written in the following form

$$
\begin{aligned}
\varphi^{(+)}(0) &= \arcsin\left(\frac{\dot{n}_1(0)}{2T\sqrt{n_1(0)n_2(0)}}\right) \equiv \phi_0, && (3.34) \\
\varphi^{(-)}(0) &= \pi - \phi_0, && (3.35)
\end{aligned}
$$

where arcsin stands for the principal value of the inverse sin. In other words, ϕ_0 defined after Eq. (3.34) is a solution to Eq. (3.33) in the interval $[-\frac{\pi}{2}, \frac{\pi}{2}]$. The existence of two solutions to the consistency conditions means that for a given density in our simple system the manifold of density-consistent initial states consists of the following two wave functions

$$
\psi_1^{(+)}(0) = \sqrt{n_1(0)}e^{i\frac{\phi_0}{2}}, \; \psi_2^{(+)}(0) = \sqrt{n_2(0)}e^{-i\frac{\phi_0}{2}} \tag{3.36}
$$

$$
\psi_1^{(-)}(0) = \sqrt{n_1(0)}e^{-i\frac{\phi_0}{2}}, \; \psi_2^{(-)}(0) = -\sqrt{n_2(0)}e^{i\frac{\phi_0}{2}} \tag{3.37}
$$

where we disregarded an irrelevant common phase factor. By substituting Eqs. (3.36) and (3.37) into the condition of Eq. (3.28) we find that the initial states $\psi^{(\pm)}(0) \in \Omega$ provided $\phi_0 \neq \pm\pi/2$. Obviously this puts a restriction on the initial values of n and $\dot{n}$. Equation (3.34) tells us that the condition $\phi_0 \neq \pm\pi/2$ actually ensures that $\dot{n}(0)$ is properly bounded [see Eqs. (3.20) and (3.21)].

It is interesting to note that if $\dot{n}(0) = 0$, than $\phi_0 = 0$ and the density-consistent initial states Eqs. (3.36) and (3.37) can be viewed as the ground (symmetric) and the excited (antisymmetric) states of a dimer in the presence of some static potential.

Now we can solve NLSE of Eq. (3.30) starting from one of the allowed initial states. The solution should return the wave function and the potential as unique functionals of the given density $n_{\mathbf{r}}(t)$. Inserting the polar representation Eq. (3.31) into Eq. (3.30) we observe that the following form of $\psi_{\mathbf{r}}(t)$,

$$
\psi_1(t) = \sqrt{n_1(t)}e^{i\varphi(t)/2}, \; \psi_2(t) = \sqrt{n_2(t)}e^{-i\varphi(t)/2}, \tag{3.38}
$$

solves NLSE if the time-dependent phase $\varphi(t)$ satisfies the equation

$$\dot{n}_1(t) = 2T\sqrt{n_1(t)n_2(t)}\sin\varphi(t). \tag{3.39}$$

For each initial state from the set of Eqs. (3.36) and (3.37) this equation has a unique solution, provided the condition $k_{12}(\psi) \neq 0$ is fulfilled.

Assume that we started from the state $\psi_1^{(+)}(0)$, Eq. (3.36). Then the solution to Eq. (3.39) reads

$$\varphi = \arcsin\left(\frac{\dot{n}_1}{2T\sqrt{n_1 n_2}}\right). \tag{3.40}$$

This equation together with Eq. (3.38) gives the wave function as a functional of the density. As long as this solution exists, the element $k_{12} = 2T\mathrm{Re}\rho_{12}$ of the $\hat{K}$ matrix stays positive

$$k_{12}^{(+)} = 2T\sqrt{n_1 n_2}\cos\varphi = \sqrt{4T^2 n_1 n_2 - \dot{n}_1^2} \tag{3.41}$$

To find the potential $v(t)$ as a functional of the density we insert Eqs. (3.38) and (3.41) into Eq. (3.29). The results takes the following form

$$v^{(+)}[n] = -\frac{\ddot{n}_1 + 2T^2(n_1 - n_2)}{2\sqrt{4T^2 n_1 n_2 - \dot{n}_1^2}}. \tag{3.42}$$

This functional reproduces the result obtained in Ref. [103]. In addition there is another solution that corresponds to another density-consistent initial state.

If we start from the second initial state, $\psi_1^{(-)}(0)$ of Eq. (3.37), we should take the second solution of Eq. (3.39) for the phase, namely

$$\varphi = \pi - \arcsin\left(\frac{\dot{n}_1}{2T\sqrt{n_1 n_2}}\right). \tag{3.43}$$

In this case k_{12} changes a sign,

$$k_{12}^{(-)} = -\sqrt{4T^2 n_1 n_2 - \dot{n}_1^2}, \tag{3.44}$$

which implies that the sign of the potential v is also reversed

$$v^{(-)}[n] = \frac{\ddot{n}_1 + 2T^2(n_1 - n_2)}{2\sqrt{4T^2 n_1 n_2 - \dot{n}_1^2}}. \tag{3.45}$$

Thus, for different initial conditions the NLSE machinery produces unambiguously different functionals $v[n]$ and $\psi[n]$. This nicely displays the initial state dependence in TDDFT [102, 120], although in the present case the dependence is very simple.

For each density-consistent initial state the unique solution to NLSE of Eq. (3.30) exists as long as the boundary of the subset Ω is not reached. This happens if $k_{12}(\psi) = 0$, i. e. when the expression under the square root in Eqs. (3.41) or (3.44) turns into zero. In agreement with the general discussion in section 3.3, at this point the bound on the time derivative of the density,

$$|\dot{n}_1| < 2T\sqrt{n_1 n_2}, \tag{3.46}$$

is saturated.

In our simple model we can also visualize and completely characterize the geometry of the v-representability subset Ω in the Hilbert space $\mathcal{H}$. Since we have an effective 2-level system the projective Hilbert space can be represented by a 2-sphere, known as a Bloch sphere. Specifically, after taking out a common phase factor, each normalized state from $\mathcal{H}$ is mapped to a point on a 2-sphere in $\mathbb{R}^3$ (see Fig. 1) by parametrizing the wave function as follows

$$|\psi\rangle = \cos\theta/2 e^{i\varphi/2} |1\rangle + \sin\theta/2 \, e^{-i\varphi/2}|2\rangle, \tag{3.47}$$

where $|1\rangle$ and $|2\rangle$ are the orthogonal states corresponding to the particle residing on sites 1 and 2, respectively. In this mapping the moduli of the on-site amplitudes are represented by the azimuthal angle θ, while the phase difference φ corresponds to the polar angle in spherical coordinates. As we can see from Fig. 1 the line $k_{12}(\psi) = 0$ divides the projective Hilbert space into two hemispheres, left and right, with $k_{12} > 0$ and $k_{12} < 0$. The two hemispheres represent two disconnected parts of the v-representability subset Ω, separated by the boundary line $k_{12} = 0$. The boundary line contains all states for which the statements of Theorem 3.3.1 do not hold. Starting from any point on those hemispheres, i. e. from a state $\psi \in \Omega$, we uniquely recover the time evolution of the system with a given density by solving NLSE of Eq. (3.30). As long as the trajectory stays within the original hemisphere and does not touch the boundary, the one-to- one density-potential map exists with the functional $v[n]$ given by Eq. (3.42) or by Eq. (3.45), depending on the hemisphere. Whether or not it is possible to construct a unique and
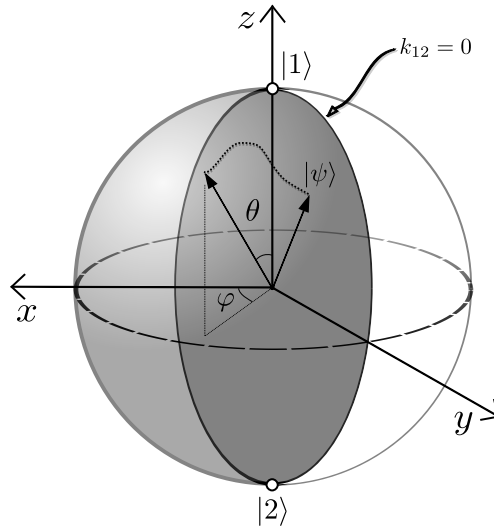
FIGURE 3.2: Each normalized state in the Hilbert space $\mathcal{H}$ maps to a point on the Bloch sphere. The north $|1\rangle$ and the south $|2\rangle$ poles correspond to the particle on sites 1 and 2. The line $k_{12} = 0$ divides the sphere into two (left and right) hemispheres corresponding to two disconnected parts of the v-representability subset Ω.

universal analytic continuation for crossing the boundary and covering the whole subset Ω is an interesting question which cannot be answered at the level of Theorem 3.3.1.

3.5 Time-dependent v-representability for a system evolving from the ground state

In this section we return to the most general case, and show that the ground state of a lattice N-particle system always belongs to the v-representability subset Ω. This implies that the lattice TDDFT is guaranteed to exist if the dynamics starts from the ground state.

Assume that $\psi_k = |k\rangle$ form a complete set of eigenstates for the lattice many-body Hamiltonian describing N-particle system in the presence of a static scalar potential $v_0(\mathbf{r})$. Let $\psi_0 = |0\rangle$ be the ground state. We are going to prove that the matrix $\hat{K}(\psi_0)$ evaluated at the ground state is strictly negative definite in the subspace of potentials

that are orthogonal to a space-constant vector V_C, i. e.,

$$V^T \hat{K}(\psi_0)V \equiv \sum_{\mathbf{r},\mathbf{r}'} v(\mathbf{r})k_{\mathbf{r},\mathbf{r}'}v(\mathbf{r}') < 0, \tag{3.48}$$

for all M-dimensional vectors $V = \{v(\mathbf{r})\}$ which satisfy the following orthogonality relation

$$V^T V_C = C\sum_{\mathbf{r}} v(\mathbf{r}) = 0, \tag{3.49}$$

where V^T stands for the transposed vector.

Our starting point is the f-sum rule (see, e. g., Ref. [121]) for the density-density response function $\chi_{\mathbf{r},\mathbf{r}'}(\omega) = \langle\langle \hat{n}_{\mathbf{r}}; \hat{n}_{\mathbf{r}'} \rangle\rangle_\omega$:

$$-\frac{2}{\pi}\int_0^\infty \omega \mathrm{Im}\chi_{\mathbf{r},\mathbf{r}'}(\omega)d\omega = i\langle 0|[\dot{\hat{n}}_{\mathbf{r}}, \hat{n}_{\mathbf{r}'}]|0\rangle. \tag{3.50}$$

To calculate the commutator in the right hand side of Eq. (3.50) we switch to the second quantized representation and write the equation of motion for the density operator $\hat{n}_{\mathbf{r}} = \hat{a}_{\mathbf{r}}^\dagger \hat{a}_{\mathbf{r}}$,

$$\frac{d\hat{n}_{\mathbf{r}}}{dt} = i\sum_{\mathbf{r}'}(T_{\mathbf{r},\mathbf{r}'}\hat{a}_{\mathbf{r}}^\dagger\hat{a}_{\mathbf{r}'} - T_{\mathbf{r}',\mathbf{r}}\hat{a}_{\mathbf{r}'}^\dagger\hat{a}_{\mathbf{r}}), \tag{3.51}$$

where $a_{\mathbf{r}}$ and $\hat{a}_{\mathbf{r}}^\dagger$ are the on-site annihilation and creation operators. Equation (3.51) is nothing but the operator form of the continuity equation. Using Eq. (3.51) one can easily calculate the commutator entering the right hand side of Eq. (3.50),

$$i[\dot{\hat{n}}_{\mathbf{r}}, \hat{n}_{\mathbf{r}'}] = -T_{\mathbf{r},\mathbf{r}'}\hat{a}_{\mathbf{r}}^\dagger\hat{a}_{\mathbf{r}'} + \delta_{\mathbf{r}\mathbf{r}'}\sum_{\mathbf{r}''}T_{\mathbf{r},\mathbf{r}''}\hat{a}_{\mathbf{r}}^\dagger\hat{a}_{\mathbf{r}''} + h.c. \tag{3.52}$$

Taking the ground state expectation value of this equation and comparing the result with Eq. (4.33) we find

$$i\langle 0|[\dot{\hat{n}}_{\mathbf{r}}, \hat{n}_{\mathbf{r}'}]|0\rangle = -k_{\mathbf{r},\mathbf{r}'}. \tag{3.53}$$

Therefore the right hand side of the lattice f-sum rule is identified with the $\hat{K}$ matrix entering the definition of the v-representability subset.[2]

---

[2]This result is not surprizing as the matrix $\hat{K}$ is a lattice analog of the Sturm-Liouville operator $m^{-1}\nabla n\nabla$, while the latter defines the right hand side of the usual f-sum rule in the contunuum (it reduces to $q^2 n/m$ for a homogeneous system).

On the other hand, for the imaginary part of density response function we have the following spectral representation [121]

$$\int_0^\infty \omega \mathrm{Im}\chi_{\mathbf{r},\mathbf{r}'}(\omega)d\omega = -2\pi\mathrm{Re}\sum_k \omega_{k0}\langle 0|\hat{n}_{\mathbf{r}}|k\rangle\langle k|\hat{n}_{\mathbf{r}'}|0\rangle, \qquad (3.54)$$

where $\omega_{k0} = E_k - E_0$ are excitation energies of the system. Substitution of Eqs. (3.53) and (3.54) into Eq. (3.50) leads to the spectral representation for the elements of $\hat{K}$ matrix

$$k_{\mathbf{r},\mathbf{r}'} = -4\mathrm{Re}\sum_k \omega_{k0}\langle 0|\hat{n}_{\mathbf{r}}|k\rangle\langle k|\hat{n}_{\mathbf{r}'}|0\rangle. \qquad (3.55)$$

Finally, inserting $k_{\mathbf{r},\mathbf{r}'}$ of Eq. (4.41) into the left hand side of Eq. (4.39), we arrive to the following remarkable result

$$\begin{aligned}V^T\hat{K}(\psi_0)V &= -4\sum_k \omega_{k0}\left|\sum_{\mathbf{r}} v(\mathbf{r})\langle 0|\hat{n}_{\mathbf{r}}|k\rangle\right|^2\\ &= -4\sum_k \omega_{k0}|\langle 0|\hat{v}|k\rangle|^2 \leq 0,\end{aligned} \qquad (3.56)$$

where $\hat{v}$ is a many-body operator corresponding to the potential $v(\mathbf{r})$,

$$\hat{v} = \sum_{\mathbf{r}} v(\mathbf{r})\hat{n}_{\mathbf{r}}. \qquad (3.57)$$

Let us show that the equality in Eq. (4.42) holds only for a space-constant potential $v_C(\mathbf{r}) = C$. Since each term in the sum in Eq. (4.42) is non-negative, the result of summation is zero if and only if

$$\langle 0|\hat{v}|k\rangle = 0, \quad \text{for all } k \neq 0. \qquad (3.58)$$

Physically the right hand side of Eq. (4.42) is proportional to the energy absorbed by a system after a small amplitude pulse of the form $v(\mathbf{r};t) = v(\mathbf{r})\delta(t)$. Then the condition Eq. (4.44) simply states that nothing is absorbed only if the potential $v(\mathbf{r})$ does not couple the ground state to any excited state.

Assume that Eq. (4.44) is fulfilled and expand the vector $\hat{v}|0\rangle$ in the complete set of states $\{|k\rangle\}$

$$\hat{v}|0\rangle = \sum_k |k\rangle\langle k|\hat{v}|0\rangle = |0\rangle\langle 0|\hat{v}|0\rangle \equiv \lambda|0\rangle. \qquad (3.59)$$

Therefore the condition of Eq. (4.44) implies that the ground state $|0\rangle$ is an eigenfunction of the operator $\hat{v}$. Since $\hat{v}$ corresponds to a local multiplicative one-particle potential, this can happen only if the potential is a constant. For clarity we write Eq. (4.45) in the coordinate representation

$$\sum_{j=1}^{N} v(\mathbf{r}_j)\psi_0(\mathbf{r}_1, ..., \mathbf{r}_N) = \lambda\psi_0(\mathbf{r}_1, ..., \mathbf{r}_N). \tag{3.60}$$

Obviously this equation can be fulfilled only if the function $v(\mathbf{r})$ takes the same value λ/N on all lattice sites, which corresponds to a space-constant potential. A notable exception is a geometrically disconnected lattice consisting of several pieces that cannot be connected by a path composed of links with nonzero $T_{\mathbf{r},\mathbf{r}'}$. In this case the arguments of the wave function $\psi_0(\mathbf{r}_1, ..., \mathbf{r}_N)$ form "disconnected groups" of coordinates corresponding to particles residing in the disconnected parts of the system. The coordinates of different disconnected groups take values in "non-overlapping" parts of the lattice. Since the number of particles in each part (number of coordinates in each group) is fixed, Eq. (3.60) can also be satisfied with a piecewise constant potential.

Therefore we arrive to the following conclusion: for a connected lattice Eq. (4.44) is fulfilled, and the inequality in Eq. (4.42) is saturated only for a constant in space potential. For all potentials which are orthogonal to a constant in a sense of Eq. (4.40) the strict inequality in Eq. (4.42) takes place. This means that matrix $\hat{K}(\psi_0)$ is negative definite and thus invertible in the $M-1$-dimensional subspace of $\mathcal{V}$ orthogonal to a constant vector V_C. In other words, the ground state of an N-particle system on a connected lattice does belong to the v-representability subset Ω. This result combined with the general existence theorem of section 3.3 proves the following particular version of the time-dependent v-representability theorem.

Theorem 3.5.1 (Ground state v-representability). Let the initial state ψ_0 for the time-dependent many-body problem on a connected lattice correspond to a ground state in the presence of some scalar potential $v_0(\mathbf{r})$. Then any density $n(\mathbf{r};t)$ which satisfies the consistency conditions of Eqs. (3.13) and (3.14) and has a continuous second time derivative is locally v-representable.

This theorem is in a clear agreement with the known statement of v-representability in the linear response regime [11].

It is worth noting that the above ground state-based argumentation can be straight-forwardly extended to a thermal equilibrium state. In this case Eq. (4.42) takes the form

$$V^T \hat{K}^\beta V = 4 \sum_{k>l} \omega_{kl} \frac{e^{-\beta E_k} - e^{-\beta E_l}}{\mathcal{Z}} |\langle l|\hat{v}|k\rangle|^2, \qquad (3.61)$$

where β is the inverse temperature, $\hat{K}^\beta$ is the $\hat{K}$ matrix evaluated for the thermal equilibrium state, $\mathcal{Z}$ is the partition function, and $\omega_{kl} = E_k - E_l$. By the same token the form defined by Eq. (3.61) is strictly negative for all potentials orthogonal to a constant vector. Therefore Theorem 2 should also apply to the ensemble version of TDDFT based on the von Neuman equation for the N-body density matrix. Of course in this case we need to prove the ensemble extension of Theorem 1, but currently this also seems relatively straightforward.

Chapter 4

Quantum electrodynamical time-dependent density-functional theory for many-electron systems on a lattice

4.1 Introduction

In the present chapter we extend the uniqueness and existence theorems of the lattice TDDFT to systems strongly interacting with quantized electromagnetic field. This generalization, which can be named as QED-TDDFT, has been proposed recently in Ref. [40].

To make the idea of the proof more transparent we start with the simplest nontrivial system of one electron on a two-site lattice (a Hubbard dimer) coupled to a single photonic mode. It is worth noting that formulation of TDDFT for this system has

its own value. Indeed, a dimer coupled to a quantum Bose field is equivalent to such well known and popular models such as the quantum Rabi model and the spin-boson model. [42–44] For this system we prove that, provided some well-defined conditions are fulfilled, there exists a unique mapping from the time-dependent on-site density and the expectation value of the bosonic coordinate to the wave function and the external driving potentials. Afterwards we extend the QED-TDDFT mapping theorem to the general case of N interacting electrons on an M-site lattice coupled to multiple photonic modes. Finally we extend theorem 3.5.1 to cover the local existence/v-representability of the dynamics started from the ground state of general lattice Hamiltonian.

This chapter is structured as the following. In section 4.2 we present a complete formulation of QED-TDDFT for the Hubbard dimer coupled to a single photonic mode. We derive the equation of motion for the expectation value of the field and the force balance equation and construct the corresponding universal (NLSE). Then we prove the mapping theorem of QED-TDDFT for this model by applying the known results from the theory of semilinear PDE [114, 115]. In section 4.3 we generalize the formalism to the system of many particles on a many-site lattice which is coupled to multiple photonic modes. We derive the corresponding NLSE to the many-body system and then formulate and prove the general existence and uniqueness theorem for the lattice QED-TDDFT. Section 4.4 presents a practically important case of a system evolving from its ground state. The main outcome of this section is the theorem of a local v-representability for the initial ground state. In Conclusion we summarize our results.

4.2 QED-TDDFT for a Hubbard dimer coupled to a single photonic mode

To make our approach more transparent and clear we first consider a simple system of one quantum particle on a two-site lattice, which is coupled to a single mode photonic field. The state of the system at time t is characterized by the electron-photon wave function $\psi_i(p; t) = \langle i, p | \Psi(t) \rangle$, where the index $i = \{1, 2\}$ corresponds to the particle "coordinate" and takes values on the lattice sites, and the real continuum variable p describes the photonic degree of freedom.
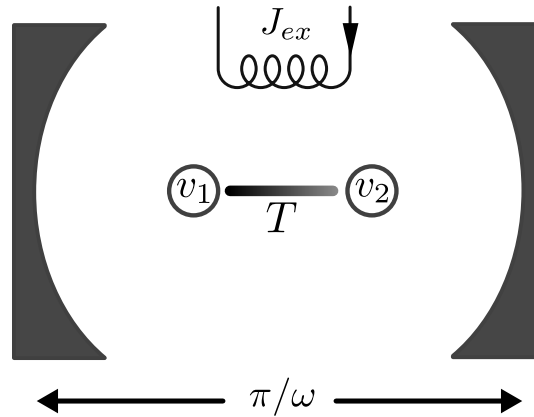
FIGURE 4.1: A schematic view of a two-site lattice in a cavity. The electron, which can tunnel from one site to the other with the hopping rate T, experiences the on-site potential $v_i(t)$ specific to that site. The photonic field in the cavity is driven by a time-dependent external dipole moment $d_{ext}(t)$. The wave length of the electromagnetic field $2\pi/\omega$ is proportional to the cavity size and assumed to be much larger than the lattice size, so that we can adopt the dipole approximation for the electron-photon interaction.

In this model the electronic density is coupled to the external on-site potential $v_i(t)$ which acts on the individual sites, while the photonic subsystem can be driven (excited) independently by an external time-dependent dipole moment $d_{ext}(t)$. Assuming that the wavelength of the photon field is much larger than the size of the system, we adopt the dipole approximation, see section 2.4. It is convenient to use the length gauge for the photon field. In this case the photon variable p is associated to the electric field which is coupled to the electron dipole moment [40, 41]. Figure 4.1 shows a schematic view of a two-site lattice in a quantum cavity.

The following time-dependent Schrödinger equation governs the time evolution of the electron-photon wave function $\psi_i(p;t)$ from a given initial state $\psi_i(p, t_0)$

$$i\partial_t\psi_1(p;t) = -T\psi_2(p;t) + \left(\frac{-\partial_p^2}{2} + \frac{\omega^2 p^2}{2} + d_{ext}(t)p + \lambda p + v_1(t)\right)\psi_1(p;t) \quad (4.1a)$$

$$i\partial_t\psi_2(p;t) = -T\psi_1(p;t) + \left(\frac{-\partial_p^2}{2} + \frac{\omega^2 p^2}{2} + d_{ext}(t)p - \lambda p + v_2(t)\right)\psi_2(p;t) \quad (4.1b)$$

where the real coefficient T corresponds to the rate of hopping from one site to the other, ω is the frequency of the photon mode and λ is the electron-photon coupling constant (see figure 4.1).

Formally Eqs. (4.1) describes a driven two-level system coupled to a quantum harmonic

oscillator. The Hamiltonian of this system is a discretized version of the general electron-photon Hamiltonian in the length gauge Eq. (2.75). Because of the gauge invariance the physics is not changed if we add a global time-dependent constant to the potential. Therefore without loss of generality we can adopt the gauge condition $v_1 + v_2 = 0$ and define the on-site potential as follows $v(t) = v_1(t) = -v_2(t)$. With this definition the Hamiltonian in Eq. (4.1) takes the form

$$\hat{H}(t) = -T\hat{\sigma}_x + v(t)\hat{\sigma}_z + \lambda p\hat{\sigma}_z - \frac{\partial_p^2}{2} + \frac{\omega^2 p^2}{2} + d_{ext}(t)p. \qquad (4.2)$$

$\hat{\sigma}_x$, and $\hat{\sigma}_z$ are the Pauli matrices, and a 2×2 unit matrix is assumed in the last three terms. The first two terms in Eq. (4.2) correspond to a two-level system (spin 1/2), while the last three terms describe a driven harmonic oscillator. Finally the third term in Eq. (4.2) is a linear coupling between the discrete and continuous variables. It is now clear the Eq. (4.1) is equivalent to the Schrödinger equation for the Rabi model or single mode spin-boson model[42–44]. Therefore the subsequent discussion and all results of this section are directly applicable to these models. We also note that a detailed derivation of Hamiltonian Eq. (4.2) for a nonrelativistic system in a cavity can be found in Ref. [41] (see Appendix E).

Now, let us turn to the formulation of QED-TDDFT. In general all DFT-like approaches assume that the state of the system is uniquely determined by a small set of basic observables, such as the density in TDDFT, the current in TDCDFT. Below we prove a theorem which generalizes the lattice-TDDFT theorem 3.3.1 to the system coupled to a quantum oscillatoric degree of freedom as defined in Eq. (4.1). Namely, we will prove that, provided that some well defined conditions are fulfilled, the electron-photon wave function $\Psi(t)$ is uniquely determined by the on-site density n_i and the expectation value of the photonic coordinate $P = \langle p \rangle$.

In our formulation we follow the NLSE approach to TDDFT[9, 13, 104–106] and adopt the same general logic as in Ref. [106]. We start with defining the basic observables for our two-site model. The first basic variable, is the on-site density n_i – the number of particles on the site i

$$n_i(t) = \int \mathrm{d}p |\psi_i(p;t)|^2. \qquad (4.3)$$

For the electron-photon system the second, photon-related variable is required. The most natural choice [40, 41] is the expectation value P of the photonic coordinate p

$$P = \int \mathrm{d}p(|\psi_1(p;t)|^2 + |\psi_2(p;t)|^2)p. \tag{4.4}$$

The next step is to find the equations which relate the basic observables, the density n_i and the field average P, to the "external potentials", the on-site potential v_i and the external dipole moment d_{ext}. Therefore we proceed by deriving the equations of motion for these two fundamental variables.

In order to derive the relevant equation of motion for n_i we calculate the time-derivative of (4.3) and then substitute the derivatives of the wave function from the Schrödinger equation (4.1). The result takes the following form

$$\dot{n}_1(t) = -2\mathrm{Im}[T\rho_{12}(t)], \tag{4.5}$$

where $\dot{n} = \partial_t n$, and $\rho_{12}(t)$ is the density matrix ,

$$\rho_{12} = \int \rho_{12}(p;t)\mathrm{d}p = \int \psi_1^*(p;t)\psi_2(p;t)\mathrm{d}p. \tag{4.6}$$

The conservation of the particles dictates that the change in the density in one site is equal to minus the change in the other site $\dot{n}_2 = 2\mathrm{Im}T\rho_{12}$. Obviously Eq. (4.5) is a lattice version of the continuity equation for site 1. Since in the left hand side of Eq. (4.5) we have the time derivative of the on-site density, the right hand side should be identified with a current flowing along the link connecting the two sites

$$J_{12}(t) = 2\mathrm{Im}[T\rho_{12}(t)]. \tag{4.7}$$

Differentiating the continuity equation (4.5) with respect to time and replacing the derivative of the wave function from the Schrödinger equation we get an equation which connects the on-site density n_i to the on-site potential v_i

$$\begin{aligned}
\ddot{n}_1(t) &= -2T\Big(\mathrm{Re}[\rho_{12}](v_1(t) - v_2(t)) \\
&\quad + T(n_1 - n_2) + 2\lambda \int \mathrm{Re}[\rho_{12}(p)]p\mathrm{d}p\Big).
\end{aligned} \tag{4.8}$$

Physically this equation can be interpreted as the (discrete) divergence of the force balance equation for the two-site model.

A special role of Eq. (4.8) for TDDFT follows from the fact that it explicitly relates the potential disbalance $v_1(t) - v_2(t)$ to the density $n_i(t)$ and its derivatives. Like before, the conservation of the particle imposes $\ddot{n}_2 = -\ddot{n}_1$. Hence, the force balance equation for $n_2(t)$ is obtained from Eq. (4.8) by changing the sign in the right hand side.

It is worth noting that the coefficient of the potential disbalance, $v_1(t) - v_2(t)$, in the force balance equation (4.8) is the kinetic energy $k_{12} = 2T\mathrm{Re}[\rho_{12}]$ therefore for the current J_{12} and kinetic energy k_{12} we have:

$$K_{12} + \mathrm{i}J_{12} = 2T\rho_{12}. \tag{4.9}$$

Importantly, Eq. (4.8) contains only the potential disbalance $v_1 - v_2$ which reflects the well known gauge redundancy of TDDFT. For a given density the force balance equation fixes the on-site potential up to a constant. In order to resolve this issue we fix the gauge by considering on-site potentials which sum up to zero $v_1 = -v_2 = v$. This can be interpreted as a switching from the whole two dimensional space of all allowed potentials to the one dimensional space of equivalence classes for physically distinct potentials.

Next, we need to derive a similar equation for P. So we differentiate (4.4) with respect to t and simplify the right hand side using the Schrödinger equation (4.1) and the result is as follows:

$$\dot{P} = \mathrm{Im}\Big[\int \Big(\psi_1^*\partial_p\psi_1 + \psi_2^*\partial_p\psi_2\Big)\mathrm{d}p\Big]. \tag{4.10}$$

For brevity we suppressed the explicit p- and t-dependence of the wave function.

By differentiating Eq. (4.10) with respect to time and again substituting the time derivatives from the Schrödinger equation (4.1) we get an equation which relates d_{ext} to P, $\ddot{P}$ and n_i

$$\ddot{P} = -\omega^2 P - \lambda(n_1 - n_2) - d_{ext}(t). \tag{4.11}$$

This equation is, in fact, the inhomogeneous Maxwell equation projected on the single photon mode [41].

In the next subsection we will use the force balance equation (4.8) and equation of motion (4.11) to analyze the existence of a TDDFT-like theory for a two-site lattice coupled to a photonic field.

Statement of the mathematical problem and the basic existence theorem

The standard TDDFT is based on the existence of a one-to-one map between the time-dependent density and the external potential. In the case of the electron-photon system the map is slightly different. Here the two basic observables, the on-site density n_i and the expectation value of the field P, are mapped to the two external fields, the on-site potential $v(t)$ and the external dipole moment $d_{ext}(t)$.

Equations (4.1), (4.3) and (4.4) uniquely determine the instantaneous wave function $\Psi(t)$, the on-site density n_i and the field average P as functionals of the initial state Ψ_0 , the on-site potential $v(t)$ and the external dipole moment $d_{ext}(t)$. This defines a unique "direct" map $\{\Psi_0, v, d_{ext}\} \to \{\Psi, n, P\}$ that is related to the standard direct quantum mechanical problem – reconstruction of the wave function $\Psi(t)$ from the initial data and the external potentials.

The TDDFT formalism for this problem relies on the existence of a unique map from the density n, the field average P and Ψ_0 to the potential v, the external dipole moment d_{ext}, and the wave function Ψ. In other words, TDDFT assumes the existence of a unique solution to the "inverse" problem of reconstructing the state $\Psi(t)$ and the potentials from the given observables and the initial data. In the following we will refer to the corresponding map $\{\Psi_0, n, P\} \to \{\Psi, v, d_{ext}\}$ as the "inverse map" to indicate that it is related to the above inverse problem. To prove the inverse map we follow the NLSE approach to TDDFT-type theories [9, 13, 104–106].

Assuming $n_i(t)$ and $P(t)$ are given functions of time, we express v and d_{ext} from the equations (4.8) and (4.11) as follows

$$v = -\frac{\ddot{n}_1 + 2T^2(n_1 - n_2) + 4T\lambda \int \mathrm{Re}[\rho_{12}(p)]p\mathrm{d}p}{4T\mathrm{Re}[\rho_{12}]}, \tag{4.12}$$

$$d_{ext} = -\ddot{P} - \omega^2 P - \lambda(n_1 - n_2), \tag{4.13}$$

where we assumed that $\mathrm{Re}[\rho_{12}] \neq 0$.

We note that at any time, including the initial time $t = t_0$, the given density has to be consistent with the wave function. At $t = t_0$ this means that the right hand side of Eq. (4.3) evaluated at the initial wave function Ψ_0 has to be equal to $n_i(t_0)$ in the left hand side. The same follows for the first derivative of the density $\dot{n}_i$ and the field average P and its first derivative $\dot{P}$. All of them should be consistent with the initial state Ψ_0 through Eqs. (4.5), (4.4) and (4.10) respectively. The consistency conditions which should be fulfilled are the following

$$n_i(t_0) = \int \mathrm{d}p |\psi_i(p; t_0)|^2, \tag{4.14a}$$

$$\dot{n}_1(t_0) = -2\mathrm{Im}[T\rho_{12}(t_0)], \tag{4.14b}$$

$$P(t_0) = \int \mathrm{d}p \; p\big(|\psi_1(p; t_0)|^2 + |\psi_2(p; t_0)|^2\big), \tag{4.14c}$$

$$\dot{P}(t_0) = \mathrm{Im}\Big[\int \mathrm{d}p \Big(\psi_1^*(p; t_0)\partial_p \psi_1(p; t_0)$$
$$+ \psi_2^*(p; t_0)\partial_p \psi_2(p; t_0)\Big)\Big]. \tag{4.14d}$$

The on-site potential v of Eq. (4.12) and the external dipole moment d_{ext} of Eq. (4.13) can be substituted as functionals of n, P and Ψ into the Schrödinger equation (4.1). The result is a universal NLSE in which the Hamiltonian is a function of the instantaneous wave function and the (given) basic variables

$$i\partial_t \Psi(t) = H[n, P, \Psi]\Psi(t). \tag{4.15}$$

Now the question of existence of a unique QED-TDDFT map $\{\Psi_0, n_i, P\} \to \{\Psi, v, d_{ext}\}$ can be mathematically formulated as the problem of existence of a unique solution to NLSE (4.15) with given $n_i(t)$, $P(t)$ and Ψ_0.

Theorem 4.2.1 (existence of QED-TDDFT for a Hubbard dimer coupled to a photonic mode). Assume that the on-site density $n_i(t)$ is a positive, continuous function of time, which has a continuous second derivative and adds up to unity, $n_1(t) + n_2(t) = 1$. Consider $P(t)$ to be a continuous function of time with a continuous second derivative. Let Ω be a subset of the Hilbert space where $\mathrm{Re}[\rho_{12}] \neq 0$. If the initial state $\Psi_0 \in \Omega$, and the consistency conditions of Eqs. (4.14) hold true, then:

(i) there is an interval around t_0 in which NLSE (4.15) has a unique solution and, therefore, there exists a unique map $\{\Psi_0, n_i, P\} \to \{\Psi, v, d_{ext}\}$.

(ii) The solutions (i. e. the QED-TDDFT map) is not global in time if at some $t^* > t_0$ the boundary of Ω is reached.

Proof. By the condition of the theorem, $\Psi_0 \in \Omega$ where $\text{Re}[\rho] \neq 0$. Therefore the on-site potential v can be expressed in terms of the density and the wave function as given by (4.12), and the Hamiltonian $\hat{H}[n, P, \Psi]$ in the universal NLSE is well defined.

Let us rewrite NLSE (4.15) in the following form

$$i\partial_t \Psi = \left(\hat{H}_0 + \hat{H}_1[n, P, \Psi] \right) \Psi, \tag{4.16}$$

where $\hat{H}_0$ is the time-independent (linear) part of the Hamiltonian,

$$\hat{H}_0 = -\frac{1}{2}\partial_p^2 + \frac{1}{2}\omega^2 p^2 + \lambda p\hat{\sigma}_z - T\hat{\sigma}_x, \tag{4.17}$$

and $\hat{H}_1$ contains all time-dependent, in particular non-linear, terms,

$$\hat{H}_1[n, P, \Psi] = d_{ext}[n, P]p + v[n, P, \Psi]\hat{\sigma}_z. \tag{4.18}$$

Here $d_{ext}[n, P]$ and $v[n, P, \Psi]$ are defined by Eqs. (4.13) and (4.12), respectively.

Since $\hat{H}_0$ is the Hamiltonian of the static shifted harmonic oscillator, it defines a continuous propagator in the Hilbert space of square integrable functions. Therefore Eq. (4.16) can be transformed to the following integral equation,

$$\Psi(t) = \quad e^{-i\hat{H}_0(t-t_0)}\Psi_0 \tag{4.19}$$
$$-i\int_{t_0}^{t} e^{-i\hat{H}_0(t-s)}\hat{H}_1[n(s), P(s), \Psi(s)]\Psi(s)\mathrm{d}s.$$

To prove the existence of solutions to this equation we can use well established theorems from the theory of quasilinear PDE. [114, 115] [1] In particular, we apply the following result. Consider an integral equation of the form,

$$u(t) = W(t, t_0)u_0 + \int_{t_0}^{t} W(t, s)K_s(u(s))\mathrm{d}s, \tag{4.20}$$

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[1] Theorem 8 page then of Harauz.

where $W(t, s)$ is a continuous linear propagator on $T = [t_0, \infty)$ and the kernel $K_t(u)$ is continuous function of time, which is locally Lipschitz in a Banach space $\mathcal{B}$. Then there is an interval $[t_0, t^*)$ where Eq. (4.20) has a unique continuous solution.

In our case we consider L^2 as the proper Banach space $\mathcal{B}$. The kernel $K_t(\Psi) = \hat{H}_1[n(t), P(t), \Psi]\Psi$ in Eq. (4.19) is continuous and Lipschitz in L^2 if $n(t)$, $\ddot{n}(t)$, $P(t)$ and $\ddot{P}(t)$ are continuous functions of time, $\Psi \in \Omega$, $n_1, n_2 > 0$, $n_1 + n_2 = 1$ and the consistency conditions Eqs. (4.14) are fulfilled. Hence if all conditions of the theorem are satisfied Eq. (4.19) has a unique solution. Moreover since in this case Ψ_0 is in the domain of H_0, $\Psi_0 \in D(H_0)$, there exists a unique differentiable (strong) solution of Eq. (4.19) which proves the statement (i) of the theorem.

The extension theorems for quasilinear PDE imply that the local solution cannot be extended beyond some maximal existence time $t^* > t_0$ only in two cases: first, at $t \to t^*$ the solution becomes unbounded or, second, at $t \to t^*$ it reaches the boundary of Ω. In our case the solution is guaranteed to be normalized and thus bounded. Therefore we are left only with the second possibility, which proves the statement (ii) and completes the proof of the theorem. $\qquad\square$

The above theorem generalizes the results of Ref. [41] where the uniqueness (but not the existence) of the map $\{\Psi_0, n_i, P\} \to \{\Psi, v, d_{ext}\}$ has been proven for analytic in time potentials using the standard Taylor expansion technique.

The Theorem 4.2.1 can be straightforwardly generalized to the case of multiple photon modes. The only difference is that $\hat{H}_0$ in Eq. (4.17) becomes the Hamiltonian of a multidimensional shifted harmonic oscillator. The rest of the proof remains unchanged. This proves the existence of QED-TDDFT for the spin-boson model in its standard form [43]. A less obvious generalization for the system of many interacting electrons on a many-site lattice is presented in the next section.

4.3 QED-TDDFT for many electrons on many-site lattices interacting with cavity photons

In the previous section we proved the QED-TDDFT existence theorem for a system of one electron on a two-site lattice coupled to a photonic mode. Below we generalize our

results to the case of N interacting electrons on an M-site lattice coupled to L photonic modes. The state of the system is described by an electron-photon wave function $\psi(\mathbf{r}_1, \cdots, \mathbf{r}_N; \{p\})$ where coordinates $\mathbf{r}_i$ of the particles $(i = 1, 2, \cdots, N)$ take values on the lattice sites and $\{p\}$ is the set of continuous coordinates describing the photonic (oscillatoric) degrees of freedom $\{p\} = \{p_1, p_2, ..., p_L\}$. Again we assume that the electronic subsystem is driven by classical on-site potentials $v(\mathbf{r}; t)$ and each photonic mode is coupled to corresponding external dipole moment $d_{ext}^\alpha(t)$. As usual, assuming that the size of the lattice is much smaller than the wave length of the photon field, we describe the electron-photon coupling at the level of the dipole approximation with $\boldsymbol{\lambda}_\alpha$ being the coupling constant to the α-photon.

The following time-dependent Schrödinger equation describes the time evolution of the system from the initial state $\psi_0(\mathbf{r}_1, \cdots, \mathbf{r}_N; \{p\})$

$$
\begin{aligned}
\mathrm{i}\partial_t \psi(\mathbf{r}_1, ..., \mathbf{r}_N; \{p\}) = & -\sum_{i=1}^N \sum_{\mathbf{x}_i} T_{\mathbf{r}_i, \mathbf{x}_i} \psi(..., \mathbf{x}_i, ...; \{p\}) \\
& + \sum_{i=1}^N v(\mathbf{r}_j; t) \psi(\mathbf{r}_1, ..., \mathbf{r}_N; \{p\}) + \sum_{j>i} w_{\mathbf{r}_i, \mathbf{r}_j} \psi(\mathbf{r}_1, ..., \mathbf{r}_N; \{p\}) \\
& + \sum_{\alpha=1}^K \left[-\frac{1}{2}\partial_p^2 + \frac{1}{2}\omega_\alpha^2 p_\alpha^2 + d_{ext}^\alpha(t) p_\alpha \right] \psi(\mathbf{r}_1, ..., \mathbf{r}_N; \{p\}) \\
& + \sum_{i=1}^N \sum_{\alpha=1}^K \boldsymbol{\lambda}_\alpha \cdot \mathbf{r}_i p_\alpha \psi(\mathbf{r}_1, ..., \mathbf{r}_N; \{p\}),
\end{aligned}
\tag{4.21}
$$

where the real coefficients $T_{\mathbf{r}, \mathbf{r}'} = T_{\mathbf{r}', \mathbf{r}}$ correspond to the rate of hopping from site $\mathbf{r}$ to site $\mathbf{r}'$ (for definiteness we set $T_{\mathbf{r}, \mathbf{r}} = 0$), and $w_{\mathbf{r}, \mathbf{r}'}$ is the potential of a pairwise electron-electron interaction.

Following the logic of section 4.2 we define the on-site density $n(\mathbf{r})$ and the expectation value of the field P_α for a mode α, which are the basic variables for the QED-TDDFT

$$
n(\mathbf{r}) = N \sum_{\mathbf{r}_2, ..., \mathbf{r}_N} \int |\psi(\mathbf{r}, \mathbf{r}_2, ..., \mathbf{r}_N; \{p\})|^2 \mathrm{d}\mathbf{p}, \tag{4.22}
$$

$$
P_\alpha = \sum_{\mathbf{r}_1, ..., \mathbf{r}_N} \int p_\alpha |\psi(\mathbf{r}, ..., \mathbf{r}_N; \{p\})|^2 \mathrm{d}\mathbf{p}, \tag{4.23}
$$

where $\mathrm{d}\mathbf{p} = \mathrm{d}p_1 \cdots \mathrm{d}p_L$.

Similar to the two-site case we derive the force balance equation by calculating the second derivative of the density (4.22) and using the Schrödinger equation (4.21) to simplify the terms with the time derivative of the wave function

$$\ddot{n}(\mathbf{r}) = 2\text{Re}\sum_{\mathbf{r}'} T_{\mathbf{r},\mathbf{r}'}\rho(\mathbf{r},\mathbf{r}')(v(\mathbf{r}';t) - v(\mathbf{r};t)) + q(\mathbf{r};t) + f(\mathbf{r};t), \tag{4.24}$$

where $q(\mathbf{r};t)$ is the lattice divergence of the internal forces

$$\begin{aligned} q(\mathbf{r};t) = \quad & - \quad 2\text{Re}\sum_{\mathbf{r}',\mathbf{r}''} T_{\mathbf{r},\mathbf{r}'}\Big[T_{\mathbf{r}',\mathbf{r}''}\rho(\mathbf{r},\mathbf{r}'') - T_{\mathbf{r},\mathbf{r}''}\rho(\mathbf{r}',\mathbf{r}'') \\ & + \quad \rho_2(\mathbf{r},\mathbf{r}'',\mathbf{r}')(w_{\mathbf{r},\mathbf{r}''} - w_{\mathbf{r}',\mathbf{r}''})\Big], \end{aligned} \tag{4.25}$$

and $f(\mathbf{r};t)$ is the force exerted on electrons from the photonic subsystem

$$f(\mathbf{r};t) = 2\text{Re}\sum_{\alpha}\sum_{\mathbf{r}'} T_{\mathbf{r},\mathbf{r}'}\boldsymbol{\lambda}_\alpha \cdot (\mathbf{r}' - \mathbf{r})\int p_\alpha \rho(\mathbf{r},\mathbf{r}';p_\alpha)\mathrm{d}p_\alpha. \tag{4.26}$$

Here $\rho(\mathbf{r},\mathbf{r}')$ is the one-particle density matrix

$$\begin{aligned} \rho(\mathbf{r},\mathbf{r}') &= \int \rho(\mathbf{r},\mathbf{r}';p_\alpha)\mathrm{d}p_\alpha \\ &= N\sum_{\mathbf{r}_2,\dots,\mathbf{r}_N}\int \psi^*(\mathbf{r},\mathbf{r}_2,\dots,\mathbf{r}_N;\{p\})\psi(\mathbf{r}',\mathbf{r}_2,\dots,\mathbf{r}_N;\{p\})\mathrm{d}\mathbf{p} \end{aligned} \tag{4.27}$$

and $\rho_2(\mathbf{r},\mathbf{r}'',\mathbf{r}')$ is the two-particle density matrix

$$\begin{aligned} \rho_2(\mathbf{r},\mathbf{r}'',\mathbf{r}') = \quad & N(N-1)\sum_{\mathbf{r}_3,\dots,\mathbf{r}_N}\int \Big[\psi^*(\mathbf{r},\mathbf{r}'',\dots,\mathbf{r}_N;\{p\}) \\ & \times \psi(\mathbf{r}',\mathbf{r}'',\dots,\mathbf{r}_N;\{p\})\Big]\mathrm{d}\mathbf{p} \end{aligned} \tag{4.28}$$

The equation of motion for the field average P_α (4.23) is derived in the same manner as in section 4.2 by calculating the second time derivative of P_α and using the Schrödinger equation (4.21),

$$\ddot{P}_\alpha = -\omega_\alpha^2 P_\alpha - d_{ext}^\alpha - \boldsymbol{\lambda}_\alpha \cdot \mathbf{d}, \tag{4.29}$$

where $\mathbf{d}$ is the total dipole moment of the N-electron system

$$\mathbf{d} = \sum_{\mathbf{r}} \mathbf{r}\, n(\mathbf{r}). \tag{4.30}$$

The existence of QED-TDDFT is equivalent to the existence of the inverse map $\{\Psi_0, n(\mathbf{r}), P_\alpha\} \rightarrow \{\Psi, v, d_{ext}^\alpha\}$. To study this map we compile the universal NLSE by expressing the on-site potential $v(\mathbf{r})$ and the external dipole moment d_{ext}^α in terms of the fundamental observables $n(\mathbf{r})$ and P_α, and the wave function Ψ.

To find the dipole moment d_{ext}^α as a functional of the field average P_α and the density $n(\mathbf{r})$ we only need to rearrange Eq. (4.29)

$$d_{ext}^\alpha = -\left(\ddot{P}_\alpha + \omega_\alpha^2 P_\alpha + \boldsymbol{\lambda}_\alpha \cdot \mathbf{d}\right). \tag{4.31}$$

The problem of finding the potential $v(\mathbf{r})$ as a functional of $n(\mathbf{r})$ and Ψ is more involved [106] as we need to solve the system of M linear equations, Eq. (4.24), for $v(\mathbf{r})$. Let us first rewrite (4.24) in a matrix form as follows

$$\hat{K}[\Psi]V = S[\ddot{n}, \Psi], \tag{4.32}$$

where $\hat{K}$ is a real symmetric $M \times M$ matrix with elements

$$k_{\mathbf{r},\mathbf{r}'}[\Psi] = 2\text{Re}\left[T_{\mathbf{r},\mathbf{r}'}\rho(\mathbf{r}, \mathbf{r}') - \delta_{\mathbf{r},\mathbf{r}'}\sum_{\mathbf{r}''} T_{\mathbf{r},\mathbf{r}''}\rho(\mathbf{r}, \mathbf{r}'')\right] \tag{4.33}$$

and V and S are M-dimensional vectors with components

$$v_{\mathbf{r}} = v(\mathbf{r}), \tag{4.34a}$$

$$s_{\mathbf{r}}[\ddot{n}, \Psi] = -\ddot{n}(\mathbf{r}) - q[\Psi](\mathbf{r}) - f[\Psi](\mathbf{r}) \quad . \tag{4.34b}$$

The problem of inverting (solving for $v(\mathbf{r})$) the force balance equation (4.32) for a general lattice, in the context of the standard electronic TDDFT, is analyzed in the previous chapter. The same argumentation regarding the properties of the matrix $\hat{K}$ is applicable in the present case. Solving Eq. (4.24) for the on-site potential $v(\mathbf{r})$ is equivalent to multiplying both sides of Eq. (4.32) by inverse of the $\hat{K}$-matrix. Therefore the matrix $\hat{K}$ must be non-degenerate. At this point it is worth noting that because of the gauge invariance $\hat{K}$ matrix (4.33) always has at least one zero eigenvalue that corresponds to a space-constant eigenvector. Therefore if $\mathcal{V}$ is the M-dimensional space of lattice potentials $v(\mathbf{r})$, then the invertibility/nondegeneracy of $\hat{K}$ should always refer to the invertibility in an $M-1$-dimensional subspace of $\mathcal{V}$, which is orthogonal to a constant

vector $v_C(\mathbf{r}) = C$. In more physical terms this means that the force balance equation (4.33) determines the self-consistent potential $v[n, \Psi](\mathbf{r})$ only up to an arbitrary constant, like Eq. (4.8) in section 4.2. Therefore in the orthogonal subspace of $\mathcal{V}$ we have

$$V = \hat{K}^{-1}S, \tag{4.35}$$

where by $\hat{K}^{-1}$ we mean inversion in the subspace of $\mathcal{V}$. Equation (4.35) is the required functional $v[n, \Psi](\mathbf{r})$ which can be used to construct the universal NLSE.

Finally, like in the two-site case (see section 4.2) the initial state Ψ_0, the density $n(\mathbf{r})$ and the field average P_α should satisfy the consistency conditions at $t = t_0$

$$n(\mathbf{r}; t_0) = N \sum_{\mathbf{r}_2, ..., \mathbf{r}_N} \int |\psi_0(\mathbf{r}, ..., \mathbf{r}_N; \{p\})|^2 \mathrm{d}\mathbf{p}, \tag{4.36a}$$

$$\dot{n}(\mathbf{r}; t_0) = -2\mathrm{Im}[\sum_{\mathbf{r}'} T_{\mathbf{r}, \mathbf{r}'} \rho_0(\mathbf{r}, \mathbf{r}')], \tag{4.36b}$$

$$P_\alpha(t_0) = \sum_{\mathbf{r}_1, ..., \mathbf{r}_N} \int p_\alpha |\psi_0(\mathbf{r}_1, ..., \mathbf{r}_N; \{p\})|^2 \mathrm{d}\mathbf{p}, \tag{4.36c}$$

$$\dot{P}_\alpha(t_0) = \mathrm{Im} \sum_{\mathbf{r}_1, ..., \mathbf{r}_N} \int \big[\psi_0^*(\mathbf{r}_1, ..., \mathbf{r}_N; \{p\})$$
$$\times \partial_{p_\alpha} \psi_0(\mathbf{r}, ..., \mathbf{r}_N; \{p\}) \big] \mathrm{d}\mathbf{p}. \tag{4.36d}$$

By substituting the on-site potential $v(\mathbf{r})$ from Eq. (4.35), and the field average P_α from Eq. (4.31) into the Schrödinger equation (4.21) we find the proper NLSE

$$\mathrm{i}\partial_t \Psi(t) = H[n, P, \Psi]\Psi(t), \tag{4.37}$$

which we use to prove the existence of the unique inverse map $\{\Psi_0, n(\mathbf{r}), P_\alpha\} \to \{\Psi, v(\mathbf{r}), d_{ext}^\alpha\}$ and thus the existence of the QED-TDDFT in a close analogy with the Theorem 4.2.1.

Theorem 4.3.1 (existence of the QED-TDDFT for lattice systems coupled to cavity photons). Assume that a given time-dependent density $n(\mathbf{r}; t)$ is nonnegative on each lattice site, sums up to the number of particles N, and has a continuous second time derivative $\ddot{n}(\mathbf{r}; t)$. Also assume that $P_\alpha(t)$ is a continuous function of t and has a continuous second time derivatives $\ddot{P}_\alpha(t)$. Let Ω be a subset of the N-particle Hilbert space $\mathcal{H}$ where the matrix $\hat{K}[\Psi]$ (4.33) has only one zero eigenvalue corresponding to the space-constant vector. If the initial state $\Psi_0 \in \Omega$, and at time t_0 the consistency conditions of Eq. (4.36) are fulfilled, then:

(i) There is a time interval around t_0 where the many-body NLSE (4.37) has a unique solution that defines the wave function $\Psi(t)$, the external potentials $v(t)$ and $d_{ext}^\alpha(t)$ as unique functionals of the density $n(\mathbf{r}; t)$, field average P_α, and the initial state Ψ_0;

(ii) The solution of item (i) is not global in time if and only if at some maximal existence time $t^* > t_0$ the boundary of Ω is reached.

The proof of this theorem goes along the same lines as the proof of Theorem 4.2.1 in section 4.2 . We transform NLSE (4.37) to a multidimensional integral equation similar to (4.19) and then apply the general existence results [114, 115] for equations of the type of Eq. (4.20) to show that the statements (i) and (ii) are in fact true. We skip the details as the procedure is mostly a straightforward repetition of the proof presented in section 4.2.

4.4 Time-dependent v-representability for a system evolving from the ground state

In this section we will show that the ground state of a quite general lattice Hamiltonian belongs to the v-representability subset Ω. This implies that the map $\{\Psi_0, n, P_\alpha\} \to \{\Psi, v, d_{ext}^\alpha\}$ is guaranteed to exist if the dynamics starts from the ground state. The main theorem of this section is a generalization of Theorem 3.5.1.

Consider the following lattice Hamiltonian of many mutually interacting electrons coupled to photonic modes

$$\hat{H}_{\alpha,\beta,\gamma} = (\hat{T} + \hat{V} + \alpha\hat{W}_{e-e}) \otimes \hat{\mathbb{I}}_{ph} + \hat{\mathbb{I}}_e \otimes \hat{H}_{ph} + \beta H_{e-ph}, \tag{4.38}$$

where $\hat{T}$ is the usual lattice operator of the kinetic energy, $\hat{V}$ corresponds to the interaction with a local external potential, $\hat{W}$ describes the electron-electron interaction, $\hat{H}_{ph}$ is the photonic Hamiltonian and H_{e-ph} is the Hamiltonian for the interaction between electrons and the photon modes, $\hat{\mathbb{I}}_{ph}$ and $\hat{\mathbb{I}}_e$ are, respectively, the unit matrices in the photonic and the electronic sectors of the Hilbert space, and α and β are real coefficients.

Here we demonstrate that the ground state of the Hamiltonian (4.38), for any $\alpha, \beta \in \mathbb{R}$ and any on-site potential, belongs to the v-representability subset Ω if all terms in

Eq. (4.38) except for $\hat{T}$ commute with the density operator $\hat{n}_\mathbf{r}$. The proof of this quite general statement closely follows the proof of the Theorem 3.5.1. Therefore, below we only briefly go through the main line of arguments.

Assume that $\Psi_k = |k\rangle$ form a complete set of eigenstates for the Hamiltonian (4.38) and let $\Psi_0 = |0\rangle$ be the ground state. We will show that the matrix $\hat{K}[\Psi_0]$ evaluated at the ground state is strictly negative definite in the subspace of potentials that are orthogonal to a space-constant vector V_C. That is,

$$V^T \hat{K}[\Psi_0] V \equiv \sum_{\mathbf{r},\mathbf{r}'} v(\mathbf{r}) k_{\mathbf{r},\mathbf{r}'} v(\mathbf{r}) < 0, \tag{4.39}$$

for all M-dimensional vectors $V = \{v(\mathbf{r})\}$ which are orthogonal to the spatially constant potential

$$V^T V_C = C \sum_{\mathbf{r}} v(\mathbf{r}) = 0, \tag{4.40}$$

where V^T stands for a transposed vector. Therefore $\hat{K}[\Psi_0]$ is nondegenerate in the subspace orthogonal to the constant potentials.

Using the f-sum rule and the spectral representation of the density-density response function (see, for example, Ref. [122]) one can represent the elements of $\hat{K}$-matrix Eq. (4.33) as follows (see chapter 3 for details)

$$k_{\mathbf{r},\mathbf{r}'}[\Psi_0] = -4\mathrm{Re} \sum_k \omega_{k0} \langle 0|\hat{n}_\mathbf{r}|k\rangle \langle k|\hat{n}_{\mathbf{r}'}|0\rangle. \tag{4.41}$$

where $\omega_{k0} = E_k - E_0$ is excitation energy of the system from the ground state to the state k.

Substituting $k_{\mathbf{r},\mathbf{r}'}$ of Eq. (4.41) into the left hand side of Eq. (4.39) we find the following result

$$\begin{aligned}
V^T \hat{K}[\Psi_0] V &= -4 \sum_k \omega_{k0} \left| \sum_\mathbf{r} v(\mathbf{r}) \langle 0|\hat{n}_\mathbf{r}|k\rangle \right|^2 \\
&= -4 \sum_k \omega_{k0} |\langle 0|\hat{v}|k\rangle|^2 \leq 0,
\end{aligned} \tag{4.42}$$

where $\hat{v}$ is an operator corresponding to the potential $v(\mathbf{r})$,

$$\hat{v} = \sum_\mathbf{r} v(\mathbf{r}) \hat{n}_\mathbf{r}. \tag{4.43}$$

The equality in Eq. (4.42) holds only for a space-constant potential $v_C(\mathbf{r}) = C$. Indeed, since each term in the sum in Eq. (4.42) is non-negative, the result of summation is zero if and only if

$$\langle 0|\hat{v}|k\rangle = 0, \quad \text{for all } k \neq 0. \tag{4.44}$$

Assuming that Eq. (4.44) is fulfilled and expanding the vector $\hat{v}|0\rangle$ in the complete set of states $\{|k\rangle\}$ we get

$$\hat{v}|0\rangle = \sum_k |k\rangle\langle k|\hat{v}|0\rangle = |0\rangle\langle 0|\hat{v}|0\rangle \equiv \lambda|0\rangle. \tag{4.45}$$

Therefore the condition of Eq. (4.44) implies that the ground state $|0\rangle$ is an eigenfunction of the operator $\hat{v}$. Since $\hat{v}$ corresponds to a local multiplicative one-particle potential this can happen only if the potential is spatially constant. Hence, for all potentials which are orthogonal to a constant in a sense of Eq. (4.40) the strict inequality in Eq. (4.42) takes place. This means that matrix $\hat{K}[\Psi_0]$ is negative definite and thus invertible in the $M - 1$-dimensional subspace of $\mathcal{V}$ orthogonal to a constant vector V_C. In other words, the ground state of N-particle system on a connected lattice does belong to the v-representability subset Ω. This result combined with the general existence theorem 4.3.1 proves the following particular version of the time-dependent v-representability theorem.

Theorem 4.4.1 (Ground state v-representability). Let the initial state Ψ_0 for a time-dependent many-body problem on a connected lattice correspond to a ground state of a Hamiltonian of the form (4.38). Consider continuous positive density $n(\mathbf{r};t)$ and field average $P(t)$ which satisfy the consistency conditions of Eqs. (4.36) and has a continuous second time derivative. Then there is a finite interval around t_0 in which $n(\mathbf{r};t)$ and $P(t)$ can be reproduced uniquely by a time evolution of Schrödinger equation (4.21) with some time-dependent on-site potential $v_i(t)$ and external dipole moment $d_{ext}(t)$.

Note that 4.4.1 is valid for any Hamiltonian of the form of Eq. (4.38) as long as all the terms in the Hamiltonian, except for the kinetic part, commute with the density operator $\hat{n}_\mathbf{r}$, and therefore, do not contribute to the $\hat{K}$-matrix. An important special case is when the initial state is the interacting many-electron ground state which is decoupled from the photonic field, $\beta = 0$. In this case the ground state is a direct product of the electronic ground state and photonic ground state. Another practically relevant case of $\alpha = \beta = 0$ corresponds to the initial state in a form of the direct product of the noninteracting

many-electron wave function (the Slater determinant) and the photonic vacuum. For all those cases the local v-representability is guarantied by the above Theorem 4.4.1.

Chapter 5

The time-dependent exchange-correlation functional for a Hubbard dimer: quantifying non-adiabatic effect

5.1 Introduction

In this chapter we exploit the possibilities of a solvable lattice model – the 2-site Hubbard model 3.4 – to address the impact of non-locality in time in the exchange correlation functional of TDDFT. Specifically, we study resonant Rabi oscillations, a prototypical example of non-linear external field driven dynamics where the population of states changes dramatically in time. We first derive the exact ground-state Hartree-exchange-correlation (Hxc) functional for the 2-site model using the Levy-Lieb constrained search [66, 110, 111]. This functional, when used in a TDDFT context with the instantaneous time-dependent density as input, constitutes the exact adiabatic approximation which can be used as a reference to quantify the role of memory effects. By carefully studying

and quantifying the dynamics produced by TDDFT with the adiabatic Hxc potential we demonstrate that it fails both quantitatively and qualitatively to describe Rabi oscillations. In the second part of this work we apply an analytic density-potential map for lattice systems given in chapter 3 [103] to derive an explicit, fully non-adiabatic exchange-correlation density functional which correctly captures all features of Rabi dynamics in the Hubbard dimer. This functional is one of the main results of this chapter.

We introduce the physics of the Rabi effect for the Hubbard dimer, showing how the dipole moment and state occupations evolve with time during the course of resonant Rabi oscillations in Section 5.2. Then in Section 5.3 we address the same problem from a TDDFT perspective. In particular we use the exact adiabatic exchange-correlation functional as a reference to quantify memory effects. In the Section 5.4 we consider the exact interacting system in a two-level approximation which allows us to derive a new approximate Hxc potential as an explicit functional of the time-dependent density. At the end of this chapter the excellent performance of this approximation is demonstrated and explained.

5.2 Rabi oscillations for two-site Hubbard model

We consider the dynamics of two electrons on a Hubbard dimer, that is, a two-site interacting Hubbard model with on-site repulsion U and hopping parameter T. The Hamiltonian of the system reads

$$\hat{H} = - T \sum_{\sigma} \left(\hat{c}_{1\sigma}^{\dagger} \hat{c}_{2\sigma} + \hat{c}_{2\sigma}^{\dagger} \hat{c}_{1\sigma} \right) + U \left(\hat{n}_{1\uparrow} \hat{n}_{1\downarrow} + \hat{n}_{2\uparrow} \hat{n}_{2\downarrow} \right)$$
$$+ \sum_{\sigma} \left(v_1(t) \hat{n}_{1\sigma} + v_2(t) \hat{n}_{2\sigma} \right), \tag{5.1}$$

where $\hat{c}_{i\sigma}^{\dagger}$ and $\hat{c}_{i\sigma}$ are creation and annihilation operators for a spin-σ electron on site i, respectively. The $\hat{n}_{i\sigma} = \hat{c}_{i\sigma}^{\dagger} \hat{c}_{i\sigma}$ are the operators for the spin-σ density at site i, and the $v_{1,2}(t)$ are time-dependent on-site potentials. We use $\hbar = e = 1$ throughout this work. Energies are given in units of the hopping parameter T. As we will see, this simple model captures most qualitative features of Rabi oscillations in interacting systems as well as the main difficulties of describing Rabi dynamics within TDDFT.

The many-body time-dependent Schrödinger equation,

$$i\partial_t|\psi(t)\rangle = H(t)|\psi(t)\rangle, \tag{5.2}$$

describes the evolution of the system from a given initial state $|\psi_0\rangle$. Since the Hamiltonian (5.1) is independent of spin, the spin structure of the wave function $|\psi(t)\rangle$ is fixed by the initial state. In the following we study the evolution from the ground state of the Hubbard dimer and therefore it is sufficient to consider only the singlet sector of our model.

In the absence of an external potential, $v_{1,2} = 0$, the stationary singlet eigenstates of the Hamiltonian (5.1) take the form

$$|g\rangle = \mathcal{N}_g\left(\hat{c}_{1\uparrow}^\dagger\hat{c}_{1\downarrow}^\dagger + \hat{c}_{2\uparrow}^\dagger\hat{c}_{2\downarrow}^\dagger + \beta_+\left(\hat{c}_{1\uparrow}^\dagger\hat{c}_{2\downarrow}^\dagger - \hat{c}_{1\downarrow}^\dagger\hat{c}_{2\uparrow}^\dagger\right)\right)|0\rangle \quad , \tag{5.3a}$$

$$|e_1\rangle = 1/\sqrt{2}\left(\hat{c}_{1\uparrow}^\dagger\hat{c}_{1\downarrow}^\dagger + \hat{c}_{2\uparrow}^\dagger\hat{c}_{2\downarrow}^\dagger\right)|0\rangle, \tag{5.3b}$$

$$|e_2\rangle = \mathcal{N}_{e_2}\left(\hat{c}_{1\uparrow}^\dagger\hat{c}_{1\downarrow}^\dagger + \hat{c}_{2\uparrow}^\dagger\hat{c}_{2\downarrow}^\dagger + \beta_-\left(\hat{c}_{1\uparrow}^\dagger\hat{c}_{2\downarrow}^\dagger - \hat{c}_{1\downarrow}^\dagger\hat{c}_{2\uparrow}^\dagger\right)\right)|0\rangle, \tag{5.3c}$$

Here $|0\rangle$ is the vacuum state, $|g\rangle$ is the ground state, and $|e_{1,2}\rangle$ are two excited singlet states. The $\mathcal{N}_{g/e_2} = (2 + 2\beta_\pm^2)^{-1/2}$ are normalization factors and the coefficients $\beta_\pm$ are defined as

$$\beta_\pm = (U \pm \sqrt{16T^2 + U^2})/4T. \tag{5.4}$$

The energy eigenvalues corresponding to the eigenstates (5.3) are

$$E_g = 2T\beta_- \ , \tag{5.5a}$$

$$E_{e_1} = U \ , \tag{5.5b}$$

$$E_{e_2} = 2T\beta_+ \ . \tag{5.5c}$$

To simplify notations, we rewrite the external potential part in Eq. (5.1) in the form

$$\sum_\sigma (v_1\hat{n}_{1\sigma} + v_2\hat{n}_{2\sigma}) = \frac{\Delta v}{2}(\hat{n}_1 - \hat{n}_2) + C(t)(\hat{n}_1 + \hat{n}_2) \tag{5.6}$$

where $\hat{n}_i = \sum_\sigma \hat{n}_{i\sigma}$ is the operator of the number of particles on site i, $\Delta v = v_1 - v_2$

is the difference of on-site potentials, and $C(t) = (v_1(t) + v_2(t))/2$. The last term in Eq. (5.6) corresponds to a spatially uniform potential. This term can be trivially gauged away and will be ignored from now on without loss of generality. Nontrivial physical effects come only from the external potential Δv which is coupled to the difference of on-site densities. The quantity $\hat{d} = \hat{n}_1 - \hat{n}_2$ can be interpreted as the dipole moment of our simplified model of a diatomic system and its expectation value $d(t) = \langle \psi(t)|\hat{d}|\psi(t)\rangle$ uniquely determines the on-site densities $n_1(t)$ and $n_2(t)$ if the total number of particles is fixed. In the following, in particular for TDDFT, we will use the dipole moment $d(t)$ as the basic "density variable".

Since the dipole moment operator $\hat{d}$ is odd under reflection (interchange of site indices), it has nonzero matrix elements only between states of different parity. In particular, $\hat{d}$ connects the ground state $|g\rangle$ of Eq. (5.3a) only to the first excited state $|e_1\rangle$

$$d_{ge} = \langle g|\hat{d}|e_1\rangle = \frac{2}{\sqrt{1 + \beta_+^2}}, \tag{5.7}$$

while the matrix element of $\hat{d}$ between the ground state and the second excited state vanishes, $\langle g|\hat{d}|e_2\rangle = 0$.

Now we are ready to discuss Rabi oscillations in the Hubbard dimer. Let us consider the evolution of the systems from its ground state $|\psi(0)\rangle = |g\rangle$ under the action of a time periodic potential

$$\Delta v(t) = 2\mathcal{E}_0 \sin(\omega t). \tag{5.8}$$

The Rabi regime of dynamics occurs when the frequency ω of a sufficiently weak driving field approaches the frequency ω_0 of the main dipole resonance. In our case this corresponds to the frequency $\omega \sim \omega_0 = E_{e_1} - E_g$ close to the energy difference between ground and first excited states, and the amplitude $\mathcal{E}_0 \ll \omega_0/d_{ge}$.

Fig. 5.1 shows resonant dynamics of the dipole moment and state populations obtained by the numerical propagation of Eq. (5.2) for a moderately strong interaction $U = T = 1$, frequency $\omega = \omega_0 = 2.56$, $d_{ge} = 1.23$, amplitude $\mathcal{E}_0 = 0.1$ and fixed electron number $N = n_1 + n_2 = 2$. We see that the populations $p_g = |\langle g|\psi\rangle|^2$ and $p_{e_1} = |\langle e_1|\psi\rangle|^2$ of the ground and the first excited state oscillate between zero and one, while the second excited state stays practically unpopulated, $p_{e_2} = |\langle e_2|\psi\rangle|^2 \approx 0$. The dipole moment shows fast oscillations at the driving frequency ω superimposed with slow oscillations

of the envelope at the Rabi frequency $\Omega_R = d_{ge}\mathcal{E}_0$. The maximal value of the dipole moment $|d_{\max}| = d_{ge} = 1.23$ is reached at 1/4 and 3/4 of the Rabi cycle when the ground and the first excited states have equal populations of 1/2.
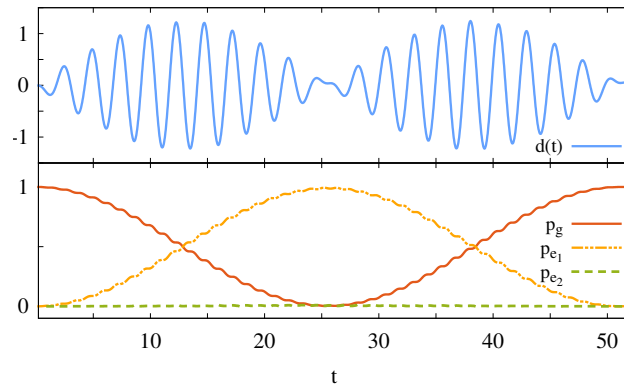
FIGURE 5.1: Rabi oscillations for resonant laser $\omega = \omega_0 = 2.56$ T . Upper panel: dipole moment $d(t)$. Lower panel: Population of ground state $p_g = |\langle g|\psi\rangle|^2$ (solid red),first excited state $p_{e_1} = |\langle e_1|\psi\rangle|^2$ (dotted orange) and second excited state $p_{e_2} = |\langle e_2|\psi\rangle|^2$ (dashed green). Time is given in units of $1/T$, where T is the hopping parameter.

The main characteristic feature of the Rabi regime is a strong variation of the state populations. It is this feature which makes the description of Rabi oscillations one of the most difficult cases for TDDFT [123, 124]. In the rest of this chapter we discuss the TDDFT approach to the Rabi dynamics for our simple two-site system.

5.3 Time-dependent Kohn-Sham equations for a Hubbard dimer

In the present two-electron case the Kohn-Sham system corresponds to two non-interacting particles which reproduce the time dependent dipole moment $d(t)$ of the interacting system. The Kohn-Sham Hamiltonian has the form of Eq. (5.1) but with no interaction ($U = 0$) and the external potential Δv^s is chosen such that the correct time-dependent density of the interacting system is reproduced.

For a singlet state both Kohn-Sham particles occupy the same one-particle Kohn-Sham orbital, which is described by two on-site amplitudes $\varphi_1(t)$ and $\varphi_2(t)$. Therefore the time-dependent Kohn-Sham equations reduce to a single 2×2 one-particle

Schrödinger equation of the form

$$i\partial_t\varphi_1 = -T\varphi_2 + \frac{\Delta v_s}{2}\varphi_1, \tag{5.9a}$$

$$i\partial_t\varphi_2 = -T\varphi_1 - \frac{\Delta v_s}{2}\varphi_2. \tag{5.9b}$$

As our dynamics starts from the ground state, Eq. (5.9) has to be solved with the initial condition $\varphi_1(0) = \varphi_2(0) = 1/\sqrt{2}$ which corresponds to the noninteracting Kohn-Sham ground state. By definition the Kohn-Sham potential $\Delta v_s(t)$ entering Eq. (5.9) produces a prescribed (interacting) dipole moment. In the present case this KS potential can be found explicitly as a functional of the density $d(t)$ using Eq. (3.29)

$$\Delta v_s[d] = -\frac{\ddot{d} + 4T^2 d}{\sqrt{4T^2(4 - d^2) - \dot{d}^2}}. \tag{5.10}$$

It is important to note that the functional $\Delta v_s[d]$ is given by Eq. (5.10) only if the system evolves from, and remains sufficiently close to, the ground state. More precisely, it is shown in section 3.4 that the functional form of Eq. (5.10) is valid as long as the condition $|\arg(\varphi_1) - \arg(\varphi_2)| < \pi/2$ is satisfied during the course of the evolution. If the opposite inequality holds, the overall sign on the right hand side of Eq. (5.10) has to be changed from $-$ to $+$. Moreover, the sign changes every time the line $|\arg(\varphi_1) - \arg(\varphi_2)| = \pi/2$ is crossed. In terms of the dipole moment, crossing this line corresponds to a vanishing expression under the square root in Eq. (5.10) [1]. The above behavior can be viewed as a manifestation of the initial state and history dependence in TDDFT [125].

The exact Kohn-Sham potential can be calculated by inserting the exact dipole moment $d(t)$ obtained from a numerical solution of the many-body Schrödinger equation (5.2) into Eq. (5.10). In order to get the Hartree-exchange-correlation (Hxc) potential we subtract the physical external potential Δv from the Kohn-Sham potential,

$$\Delta v_{\text{Hxc}} = \Delta v_s - \Delta v. \tag{5.11}$$

The time dependence of the exact Δv_{Hxc} which corresponds to the dipole moment $d(t)$ presented in Fig. 1 (i. e., to the regime of resonant Rabi oscillations, described in Sec. II)

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[1]Since the whole right hand side in this equation is proportional to the time derivative of the denominator, turning the square root into zero does not lead to a divergence because the numerator vanishes at the same time. Also, for any physical evolution the expression under the square root always stays nonnegative as $|\dot{d}|$ for a lattice is properly bounded from above (3.21).

is shown in the top panel of Fig. 5.3.

In practice the exact Hxc functional is unknown and one has to rely on approximations. The simplest and the most common approximation in TDDFT is based on the adiabatic assumption for exchange-correlation effects. Below, we present and test the adiabatic approximation for our model system.

5.3.1 Adiabatically exact functional

To construct the adiabatic approximation for the Hubbard dimer we first find the exact ground-state Hxc functional by the Levy-Lieb constrained search, i.e., we perform an exhaustive search over the space of all allowed two-particle wave functions Ψ that yield a given dipole moment d to find the Hohenberg-Kohn energy functional $F_{\mathrm{HK}}[d]$,

$$F_{\mathrm{HK}}[d] = \min_{\Psi \to d} \langle \Psi | \hat{T} + \hat{U} | \Psi \rangle, \tag{5.12}$$

where $\hat{T}$ and $\hat{U}$ are operators of the kinetic energy and the interaction energy, i. e., the first and the second terms in the Hamiltonian (5.1), respectively.

The exact ground state Hxc potential is given by the derivative of the Hxc energy with respect to the dipole moment,

$$\Delta v_{\mathrm{Hxc}}^{gs}[d] = 2 \, \frac{\partial}{\partial d} \big(F_{\mathrm{HK}}[d] - T^s[d] \big) \tag{5.13}$$

where $T^s[d]$ is the kinetic energy functional that is defined by Eq. (5.12) with $U = 0$. More details on this construction can be found in the Appendix C.

In the *adiabatically-exact* approximation the exact ground-state Hxc potential of Eq. (5.13) is used in the time-dependent Kohn-Sham equations, i.e., the Hxc potential at time t is calculated by inserting the instantaneous value of $d(t)$ into the ground-state functional

$$\Delta v_{\mathrm{Hxc}}^{ad}[d](t) = \Delta v_{\mathrm{Hxc}}^{gs}[d(t)]. \tag{5.14}$$

We note that the adiabatically-exact Hxc potential has also been found numerically for real-space one-dimensional two-electron systems in Refs. [59, 126, 127] using the iterative procedure introduced in Ref. [126]. A big advantage of the present simple model is that

we know explicitly the functional dependence of the exact ground-state Hxc potential (see Appendix C), i. e., we do not need any prior knowledge of the time-dependent density.

To test the performance of the adiabatically-exact functional in the regime of Rabi oscillations we propagate self-consistently the Kohn-Sham equations with $v_{\text{Hxc}}^{ad}[d](t)$ for the same parameters as in Sec. 5.2.
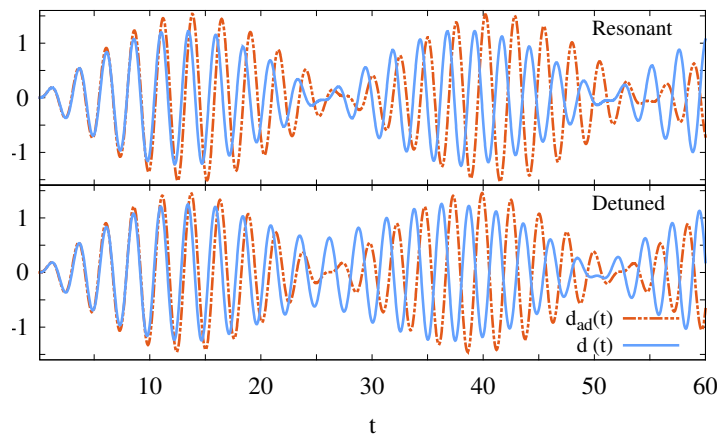
FIGURE 5.2: Upper panel : $d(t)$ (solid blue) in the presence of a laser of frequency $\omega = \omega_0 = 2.56$ T, compared to $d_{\text{ad}}(t)$ (dotted red) propagated using the exact ground state functional $\Delta v_{\text{Hxc}}^{gs}[d]$ in the presence of a laser resonant with the adiabatically exact linear response frequency $\omega_{\text{ad}}^{LR} = 2.60$ T. Lower panel: $d(t)$ for slightly detuned laser $\omega = \omega_0 + 0.03$ (solid blue) compared to $d_{\text{ad}}(t)$ using $\omega = \omega_{\text{ad}}^{LR} + 0.03$ (dotted red). Time is given in units of the inverse of the hopping parameter T.

The results of the propagation confirm a general conclusion of Ref. [123] about the presence of an artificial dynamical detuning in the description of Rabi oscillations using adiabatic functionals. In Fig. 5.2 we compare the evolution of the exact dipole moment $d(t)$ (blue) with the dipole moment $d_{\text{ad}}(t)$ (red) obtained from Kohn-Sham equations with the adiabatically-exact Hxc potential for resonant (upper panel) and slightly detuned (lower panel) applied lasers. The upper panel shows the dynamics at resonant conditions when the frequency ω of the driving field is equal to the frequency ω_{res} of the main dipole resonance. In the exact interacting system this frequency is obviously $\omega_{\text{res}} = \omega_0$, while in the approximate TDDFT it is approximation-dependent, and should be determined consistently as the frequency $\omega_{\text{res}} = \omega_{LR}$ of the corresponding linear response resonance. At the first sight, the function $d_{\text{ad}}(t)$ resulting from TDDFT with the adiabatic Hxc potential (upper panel in red on Fig. 5.2) looks qualitatively similar to the

exact $d(t)$ (upper panel in blue on Fig. 5.2). However, there is a deep difference in the underlying microscopic dynamics. The physical system returns to its initial state after two periods of the dipole moment's envelope, which corresponds to the Rabi period of $T_{\rm R} = \frac{2\pi}{\mathcal{E}_0 d_{ge}} = 51.10$. In contrast, the microscopic period of the Kohn-Sham system with the adiabatic Hxc potential coincides with that of the dipole moment, which is the characteristic feature of detuned Rabi oscillations. In fact, the Kohn-Sham Rabi dynamics is always internally detuned by the presence of the adiabatic potential $\Delta v_{\rm Hxc}^{gs}[d(t)]$ which depends on the instantaneous density [123]. While this important difference is hidden in the case of resonant dynamics of the dipole moment, it is revealed immediately when the driving frequency is a bit shifted (detuned) from the exact resonance. The dipole moments $d(t)$ and $d_{\rm ad}(t)$ for a slightly detuned driving field with $\omega = \omega_{\rm res} + 0.03$ are presented in the lower panel on Fig. 5.2. The exact dipole moment $d(t)$ develops a "neck" at $t \sim T_{\rm R}$ showing that the actual physical period is indeed $T_{\rm R} \approx 50$. On the other hand, the function $d_{\rm ad}(t)$ is practically unaffected by the external detuning because the Kohn-Sham system, being already strongly detuned internally, is insensitive to small external variations of the driving frequency. This qualitative failure of the adiabatic approximation clearly demonstrates the important role of exchange-correlation memory effects in the correct description of Rabi oscillations.

To further quantify non-adiabatic effects in the Rabi regime we extract a non-adiabatic contribution to the total Hxc potential. Namely, we subtract the adiabatic potential $\Delta v_{\rm Hxc}^{gs}[d(t)]$ evaluated at the exact dipole moment $d(t)$ from the exact $\Delta v_{\rm Hxc}(t)$ defined by Eqs. (5.11) and (5.10). In Fig. 5.3 we present the non-adiabatic part of Hxc potential together with the exact $\Delta v_{\rm Hxc}(t)$. The non-adiabatic contribution to $\Delta v_{\rm Hxc}(t)$ turns out to be more than double the amplitude of the external potential and in fact as large as the Hxc potential itself during a significant part of the Rabi-cycle. Not surprisingly, the non-adiabatic effects are small at the beginning and at the end of the Rabi cycle when the ground-state population is dominant and the system is close to the linear response regime. But they grow fast when the system is driven away from the ground-state and they remain large for a large part of the Rabi-cycle. It is interesting to notice that, centered around $T_R/2$, there is a long period of time during which the amplitude of the adiabatic effects remains almost constant (see Fig. 5.3).

Apparently a better approximation for the exchange-correlation potential is needed to capture non-adiabatic effects relevant to describe Rabi oscillations. In the next section
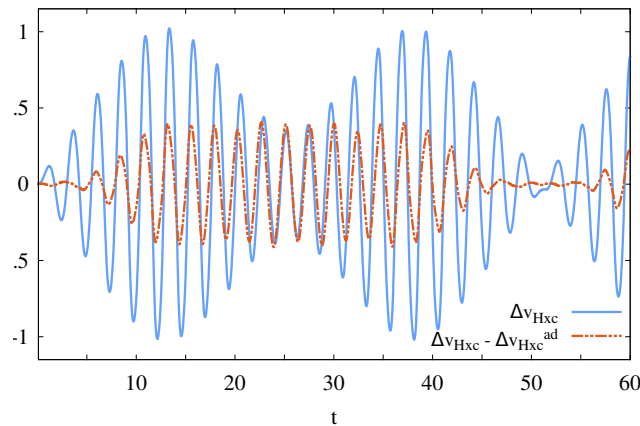
FIGURE 5.3: Time-dependent Hxc potential $\Delta v_{\text{Hxc}}(t)$ (in units of the hopping parameter T) (solid blue) and its non-adiabatic contribution defined as $\Delta v_{\text{Hxc}}(t) - \Delta v_{\text{Hxc}}^{gs}[d(t)]$ (in units of the hopping parameter T) (dotted red). Time is given in units of $1/T$.

we propose an explicit non-adiabatic density functional based on a two-level description of the interacting system.

5.4 Time-dependent exchange-correlation potential in the two-level approximation

In general, the Hxc functional $\Delta v_{\text{Hxc}}[d]$ can be found via Eq. (5.11) if we know the external potential as a functional of $d(t)$. The presence of interactions makes the problem of finding the functional $\Delta v[d]$ highly nontrivial even in our simple model. Fortunately in some cases like the Rabi oscillations the problem is simplified dramatically because the behavior of the system is close to that of an effective two-level system.

Let us assume that the second excited state $|e_2\rangle$ is not participating in the dynamics. We write the many-body Schrödinger equation (5.2) in the two level approximation as

$$i\partial_t \psi_g(t) = E_g \psi_g(t) + d_{ge} \frac{\Delta v(t)}{2} \psi_e(t) , \tag{5.15a}$$

$$i\partial_t \psi_e(t) = E_{e_1} \psi_e(t) + d_{ge} \frac{\Delta v(t)}{2} \psi_g(t) , \tag{5.15b}$$

where $\psi_g(t) = \langle g|\psi(t)\rangle$, and $\psi_e(t) = \langle e_1|\psi(t)\rangle$ are the projections of the time-dependent wave function onto the ground and first excited state, respectively.

By rotating the basis we can represent Eq. (5.15) in the form of a Schrödinger equation for one particle on an effective "two-site lattice". In other words, Eq. (5.15) is unitarily

equivalent to Eq. (5.9) with hopping constant $\omega_0/2$ and external potential given by $d_{ge}\Delta v/2$. Using this mapping and the Kohn-Sham potential Δv_s of Eq. (5.10) we can immediately write the external potential Δv of the interacting system as a functional of the dipole moment d

$$\Delta v^{2L}[d] = \frac{(-1)^p}{d_{ge}}\left(\frac{\ddot{d} + \omega_0^2 d}{\sqrt{\omega_0^2(d_{ge}^2 - d^2) - \dot{d}^2}}\right), \tag{5.16}$$

where the integer p counts how many times the square root turns into zero during the evolution. The factor $(-1)^p$ accounts for the sign changes explained after Eq. (5.10).

In order to find the Hxc potential $\Delta v_{\text{Hxc}}[d]$ as a functional of the dipole moment we substitute the external potential Δv of Eq. (5.16) and the Kohn-Sham potential of Eq. (5.10) into Eq. (5.11):

$$\Delta v_{\text{Hxc}}^{2L}[d] = -\frac{\ddot{d} + 4T^2 d}{\sqrt{4T^2(4 - d^2) - \dot{d}^2}}$$
$$-\frac{(-1)^p}{d_{ge}}\left(\frac{\ddot{d} + \omega_0^2 d}{\sqrt{\omega_0^2(d_{ge}^2 - d^2) - \dot{d}^2}}\right) \tag{5.17}$$

This expression is one of the main results of this chapter. It provides us with an explicit fully non-adiabatic density functional which, by construction, should correctly describe the Rabi oscillations. It is worth emphasizing that the functional Eq. (5.17) contains history dependence via the integer p in the second term.

One can easily check that the non-linear functional $\Delta v_{\text{Hxc}}^{2L}$ of Eq. (5.17) produces the exact dynamic exchange-correlation kernel in the linear response regime. The formally exact Hxc functional in the linear response can be written as follows

$$\Delta v_{\text{Hxc}}^{LR}[d](\omega) = \left[\chi_s^{-1}(\omega) - \chi^{-1}(\omega)\right]d = f_{xc}(\omega)d, \tag{5.18}$$

where $\chi_s(\omega)$ and $\chi(\omega)$ are the density response functions for the Kohn-Sham and the interactiving system, respectively. By definition the term in parentheses is the exact exchange-correlation kernel $f_{xc}(\omega)$. Since the eigenfunctions for the Hubbard dimer are known, Eqs. (5.3) and (5.5), we can write the exact response functions $\chi(\omega)$ in the Lehmann representation [122], and substitute it into Eq. (5.18). The result takes the

following form

$$\Delta v_{\text{Hxc}}^{LR}[d](\omega) = \left(T - \frac{\omega_0}{2d_{ge}^2} + \left(\frac{1}{4T} - \frac{1}{2\omega_0 d_{ge}^2}\right)\omega^2\right)d. \tag{5.19}$$

Now it is straightforward to see that this equation is identical to the linearized version of the approximated functional $\Delta v_{\text{Hxc}}^{2L}[d]$ defined by Eq. (5.17) with $p = 0$. In other words our approximation becomes exact in the linear regime. This nice property is not accidental because the functional of Eq. (5.17) is based on the two-level approximation. In the linear response regime, the symmetric Hubbard dimer becomes an effective two-level system because the dipole transition matrix element between the ground state $|g\rangle$ and the second excited state $|e_2\rangle$ vanishes.

A subtle property of the non-adiabatic functional Eq. (5.17) is the dependence on the second time derivative $\ddot{d}$ of the dipole moment. The presence of $\ddot{d}$ does not mean that the exchange-correlation potential assumes a dependence on the future. In general the existence theorem 3.3.1 requires the second time derivative of the density to be continuous. Therefore $\ddot{d}$ can be calculated as a left limit for any time greater than the initial time, $t > 0$. At $t = 0$ the value of $\ddot{d}(0)$ is determined by the initial value of the external potential as follows

$$\ddot{d}(0) = -d_{de}\Delta v(0)\sqrt{\omega_0^2\left(d_{ge}^2 - d^2(0)\right) - \dot{d}^2(0)} - \omega_0^2 d(0), \tag{5.20}$$

where the $d(0)$ and $\dot{d}(0)$ are fixed by the Kohn-Sham -initial state

$$d(0) = 2(|\varphi_1(0)|^2 - |\varphi_2(0)|^2), \tag{5.21a}$$

$$\dot{d}(0) = -4T\text{Im}[\varphi_1^*(0)\varphi_2(0)]. \tag{5.21b}$$

After this preliminaries we can plug the Hxc potential Eq. (5.17) into the Kohn-Sham equations and propagate them self-consistently to test the performance of our non-adiabatic approximation. It is, however, clear that the functional $\Delta v_{\text{Hxc}}^{2L}[d]$, by construction, should exactly reproduce the results of the two-level approximation to the full interacting problem. Therefore TDDFT with the Hxc potential of Eq. (5.17) is as accurate as the two-level approximation itself.

In Fig. 5.4 we compare the exact resonant Rabi dynamics of the dipole moment with
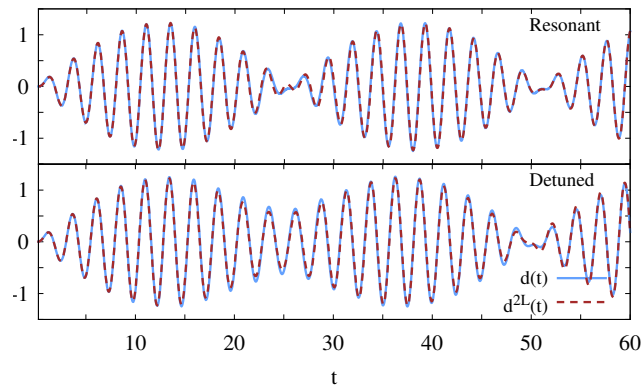
FIGURE 5.4: Upper panel: $d(t)$ (solid blue) for resonant laser frequency $\omega = \omega_0 = 2.56$ T compared to two-level approximation $d^{2L}(t)$ (dashed brown) using $\Delta v_{\text{Hxc}}^{2L}$, Eq. (5.17), and same laser frequency. Lower panel: $d(t)$ (solid blue) and $d^{2L}(t)$ (dashed red) for slightly detuned laser $\omega = \omega_0 + 0.03$ T (same detuning as lower panel on Fig. 5.2). Time is given in units of $1/T$.

the one obtained in the two-level approximation or, alternatively, by solving the Kohn-Sham equations self-consistently with the potential $\Delta v_{\text{Hxc}}^{2L}[d]$. The exact and approximate dipole moments are practically on top of each other. The non-adiabatic functional of Eq. (5.17) excellently reproduces Rabi oscillations for a resonant excitation. Apparently it also works perfectly for detuned Rabi dynamics provided that the detuning is not too large. Another nice property of this approximation is that it becomes essentially exact for a sufficiently weak non-resonant driving potential which corresponds to the linear response regime.

Chapter 6

Collective variables and time-dependent solvable systems, Using TD(C)DFT to control quantum systems

6.1   Introduction

In this chapter we use the explicit TDCDFT maps to construct analytic control signals driving a system in such a way that the prescribed behavior of the basic collective variable, the current and/or the density, is reproduced. The time dependence of the control signal and the dynamics of the wave function are then parametrized in terms of the physically intuitive observables. The analytic TDCDFT maps are known both for a particle in the real continuum space and for discrete lattice (e. g. tight binding) systems (3.1). This allows us to address, within a common scheme, control problems for the real space dynamics and for dynamics of discrete systems with a finite dimensional Hilbert space, such as a motion of quantum particle on tight-binding lattices, or the dynamics of a spin in the presence of a time-dependent magnetic field. To illustrate our strategy of inverse engineering we will recover the known exact solution for a driven harmonic

oscillator [73, 74], and present nontrivial examples of analytic control for a particle on a finite 1D chain and for a spin-1/2 (qubit) system.

The structure of this chapter is the following. In section 6.2 we present the general idea of reconstructing driving potentials for solvable problems using analytic TD(C)DFT maps. In section 6.3 we use TDCDFT to construct solvable problems for the real space one-particle dynamics. As a particular example we reconstruct the potential and the wave function generated by a density evolution in the form of a time-dependent rescaling of some initial distribution supplemented with a rigid shift in space. The corresponding solution recovers the one for the driven harmonic oscillator [73, 74]. In section 6.4 the formalism for discrete spaces is presented. In the first subsection we give an explicit example for controlling motion of a particle on atomic chain. In the second subsection the formalism is allied to a spin-1/2 control that is isomorphic to the control problem for a particle on a two-site lattice.

6.2 Construction of solvable problems via TDDFT maps: The basic idea

In the standard "direct" statement of a quantum mechanical problem the Schrödinger equation determines evolution of the wave function $\psi(t)$ from the initial state ψ_0 in the presence of a given time-dependent external potential. Thus for a given initial state the Schrödinger equation generates a map $\mathcal{V} \mapsto \mathcal{H}$ from the space $\mathcal{V}$ of external potentials to the Hilbert space $\mathcal{H}$. This direct map is shown schematically on Fig. 6.1a. Unfortunately, analytically solvable time-dependent quantum problems are exceptionally rare and to solve the time-dependent Schrödinger equation even for simplest two-level systems practically always requires numerical calculations.

To understand how TDDFT can help in finding analytically solvable problems we analyze mapping between different sets of object entering this approach. All TDDFT-type theories rely on the existence of a unique solution to a special "inverse" quantum problem. That is a possibility to uniquely reconstruct the driving field from a given evolution of the conjugated observable (such as, the density in TDDFT or the current in TDCDFT) and a given initial state. In other words, if $\mathcal{N}$ is the space of basic observables, then the existence of TDDFT implies that for a given initial state there exist two unique maps
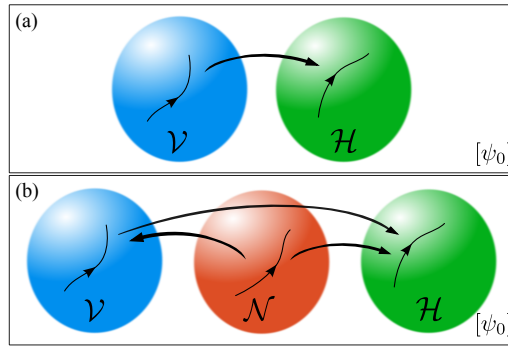
FIGURE 6.1: (a) The direct map from a trajectory in the space $\mathcal{V}$ of potentials to the trajectory in the Hilbert space $\mathcal{H}$, generated by the time-dependent Schrödinger equation for a given initial state ψ_0. (b) TDDFT mappings between trajectories in the space $\mathcal{N}$ of observables (densities or currents), space $\mathcal{V}$ of potentials and the Hilbert space $\mathcal{H}$. For a given initial state, by choosing a desired time evolution of the density/current we can reconstruct the driving potential and the wave function.

$\mathcal{N} \mapsto \mathcal{V}$ and $\mathcal{N} \mapsto \mathcal{H}$ which relate a given trajectory in the space $\mathcal{N}$ of observables to the corresponding trajectories in $\mathcal{V}$ and in $\mathcal{H}$. The composition of these TDDFT maps recovers the usual direct map $\mathcal{V} \mapsto \mathcal{H}$ generated by the time-dependent Schrödinger equation .

In general for many-particle systems the solution of the inverse problem is even more difficult than the solution of the usual Schrödinger equation . In fact, mathematically construction of the TDDFT maps is equivalent to solving a certain nonlinear quantum many-body problem [104]. However, there are special situations when the inverse problem possesses a simple analytic solution. These situations cover, in particular, generic driven one-particle dynamics both in the real space and on lattices with some mild restrictions on allowed initial states and the behavior of observables. For those cases, the TD(C)DFT maps $\mathcal{N} \mapsto \mathcal{V}$ and $\mathcal{N} \mapsto \mathcal{H}$ can be found explicitly in the analytic form [102–106]. For example, in the case of a particle driven by a time-dependent vector potential $\mathbf{A}(\mathbf{t})$ the wave function and the vector potential are the explicit analytic functionals of the current density $\mathbf{j}(\mathbf{r}, t)$ and the initial state, that is $\mathbf{A}[\mathbf{j}, \psi_0]$ and $\psi[\mathbf{j}, \psi_0]$. By construction the wave function and the potential, obtained in such a way, are connected by the Schrödinger equation with the proper initial condition. Hence by assuming different space-time distributions of the observable we can generate infinitely many solutions to the Schrödinger equation , where the potential in the Hamiltonian and the solution are expressed analytically in terms of the prescribed observable. In this setup the space of the observables plays the role of the parameter's space, while the TD(C)DFT maps

provide us with the analytic parametrization formulas for the Hamiltonian and the solution.

In the next sections we present the explicit framework for designing solvable one-particle problems in continuous and discrete spaces and illustrate our strategy of inverse engineering with several nontrivial examples.

6.3 Reconstruction of the real space potentials

In this section we illustrate our strategy of generating the exact solutions for a simpler and more familiar case of a single quantum particle in the real space. Let us consider an electron in the three-dimensional space subjected to a time-dependent external electromagnetic field. It is convenient to use the temporal gauge in which the electric $\mathbf{E}$ and magnetic $\mathbf{B}$ fields are related to the vector potential $\mathbf{A}(\mathbf{r}, t)$ as follows [1]

$$\mathbf{B} = \nabla \times \mathbf{A}; \quad \mathbf{E} = -\partial_t \mathbf{A}. \tag{6.1}$$

Given the vector potential $\mathbf{A}(\mathbf{r}, t)$ and the initial state $\psi_0(\mathbf{r}) = |\psi_0(\mathbf{r})|e^{\varphi_0(\mathbf{r})}$ the wave function $\psi(\mathbf{r}, t)$ is obtained by solving the following Schrödinger equation

$$\mathrm{i}\partial_t \psi(\mathbf{r}, t) = \frac{1}{2m}\big(-i\nabla - \mathbf{A}(\mathbf{r}, t)\big)^2 \psi(\mathbf{r}, t). \tag{6.2}$$

Here we are interested in the vector potentials for which the Schrödinger equation possesses an analytic solution. Therefore we follow our strategy and apply TDCDFT maps $\mathbf{j} \mapsto \mathbf{A}$ and $\mathbf{j} \mapsto \psi$ from the time-dependent current $\mathbf{j}(\mathbf{r}, t)$ to the vector potential and the wave function. These maps can be easily found explicitly provided the initial state is nodeless $\psi_0(\mathbf{r}) \neq 0$, and the current $\mathbf{j}(\mathbf{r}, t)$ fulfills the condition

$$\int_{t_0}^{t} \nabla \cdot \mathbf{j}(\mathbf{r}, t) \neq |\psi_0(\mathbf{r})|^2 = n_0(\mathbf{r}), \tag{6.3}$$

which ensures the state to remain nodeless in the course of evolution. Then $\mathbf{A}(\mathbf{r}, t)$ and the time-dependent wave function $\psi(\mathbf{r}, t)$ are uniquely reconstructed from the given

---

[1] Throughout this article we work in the system of units in which $\hbar, c, e = 1$.

current [104]

$$A[\psi_0, \mathbf{j}] = \nabla\varphi(\mathbf{r}, t) - m\mathbf{v}(\mathbf{r}, t), \tag{6.4a}$$

$$\psi[\psi_0, \mathbf{j}] = \sqrt{n(\mathbf{r}, t)}e^{i\varphi(\mathbf{r}, t)}, \tag{6.4b}$$

$$\varphi[\psi_0, \mathbf{j}] = \varphi_0 + \int_{t_0}^{t} dt' \left(\frac{\nabla^2 \sqrt{n(\mathbf{r}, t')}}{2m\sqrt{n(\mathbf{r}, t')}} - \frac{1}{2}m\mathbf{v}^2(\mathbf{r}, t) \right). \tag{6.4c}$$

where the density $n(\mathbf{r}, t)$ and the velocity $\mathbf{v}(\mathbf{r}, t)$ are defined as follows

$$n(\mathbf{r}, t) = -\int_{t_0}^{t} \nabla \cdot \mathbf{j}(\mathbf{r}, t) + n_0(\mathbf{r}), \tag{6.5a}$$

$$\mathbf{v}(\mathbf{r}, t) = \frac{\mathbf{j}(\mathbf{r}, t)}{n(\mathbf{r}, t)}. \tag{6.5b}$$

One can straightforwardly check, by a direct substitution, that these formulas indeed give a solution to Eq. (6.2). These equations allow us to reconstruct the driving potential for a prescribed evolution of the current.

Using Eqs. (6.4) with different time-dependent currents we can construct infinitely many Schrödinger equations with time-dependent vector potentials, which are all analytically solvable and the solutions are given by Eqs. (6.4b) and (6.4c).

Now we turn to a more specific situation when the system is driven by a longitudinal electric field $\mathbf{E}$ at zero magnetic field, $\mathbf{B} = 0$. In the absence of the magnetic field $\nabla \times \mathbf{A} = 0$ and therefore Eq. (6.4a) implies that the curl of the velocity also vanishes, $\nabla \times \mathbf{v} = 0$. The velocity of a one-particle system driven by a potential field must be potential. In this case it is natural to use the Coulomb gauge ($\nabla \cdot \mathbf{A} = 0$) and express the electric field as a gradient of the scalar potential $V(\mathbf{r}, t)$, that is $\mathbf{E} = -\nabla V$. By applying the standard gauge transformation to Eq. (6.2) we obtain the Schrödinger equation of the following form

$$i\partial_t \psi(\mathbf{r}, t) = \left(-\frac{\nabla^2}{2m} + V(\mathbf{r}, t) \right) \psi(\mathbf{r}, t). \tag{6.6}$$

where $\psi(\mathbf{r}, t)$ is now the time-dependent wave function in the Coulomb gauge.

Applying the gauge transformation Eqs. (6.4) we find the mapping from the current $\mathbf{j}(\mathbf{r}, t)$ or equivalently the velocity and density $n(\mathbf{r}, t)$ to the external potential $V(\mathbf{r}, t)$

and the wave function $\psi(\mathbf{r}, t)$ [102]

$$
\begin{aligned}
V(\mathbf{r}, t) &= \frac{\nabla^2 \sqrt{n(\mathbf{r}, t)}}{2m\sqrt{n(\mathbf{r}, t)}} - m \int_0^{\mathbf{r}} \dot{\mathbf{v}}(\mathbf{r}', t) \cdot \mathrm{d}\mathbf{r}' \\
&\quad - \frac{1}{2} m \mathbf{v}^2 - \dot{C}(t) \tag{6.7a} \\
\psi(\mathbf{r}, t) &= \sqrt{n(\mathbf{r}, t)} e^{i\tilde{\phi}(\mathbf{r}, t)}, \tag{6.7b} \\
\tilde{\phi}(\mathbf{r}, t) &= \int_0^{\mathbf{r}} m\mathbf{v}(\mathbf{r}', t) \cdot \mathrm{d}\mathbf{r}' + C(t), \tag{6.7c}
\end{aligned}
$$

where $C(t)$ is a time-dependent constant. Since the velocity is irrotational by construction the line integrals in Eqs. (6.7a) and (6.7c) do not depend on the integration path. Therefore we indicate only the initial and the final points of the path. The value $C(t_0)$ at the initial time is uniquely determined by the initial condition, while for $t > t_0$ the function $C(t)$ is arbitrary and can be chosen at convenience, for example to fix the value of the potential at infinity. The presence of a time-dependent constant in the density-potential mapping is in agreement with the Runge-Gross theorem [6]. The first term in Eq. (6.7a) is the Bohm potential that can be physically interpreted as an adiabatic potential for which the prescribed (nodeless) $n(\mathbf{r}, t)$ is the instantaneous ground state density. The second and the third terms in Eq. (6.7a) are related to inertia forces. These terms compensate the inertia forces exerted on a particle in a local non-inertial frame moving with the velocity $\mathbf{v}(\mathbf{r}, t)$. As a result in this co-moving frame the density stays stationary and equal to the initial density distribution. In the original frame the velocity-dependent contribution appears as a deformation of the adiabatic potential, which is aimed to support the prescribed density in the case of arbitrary fast evolution.

There is an important difference of the present construction and the explicit current-vector potential mapping of Eqs. (6.4)-(6.5b). Eqs. (6.7) state that given the density n and the corresponding velocity $\mathbf{v}$, the external potential V and the wave function ψ can be found analytically. However the density and the velocity are not independent variables as they have to be consistent through the continuity equation

$$
\dot{n}(\mathbf{r}, t) = -\nabla \cdot [n(\mathbf{r}, t)\mathbf{v}(\mathbf{r}, t)] . \tag{6.8}
$$

The complication comes from the requirement of irrotational velocity, $\nabla \times \mathbf{v} = 0$ which implies the velocity field of the form $\mathbf{v} = \nabla \Phi$. Because of this condition there is no simple and universal analytic relation between the observables entering equation (6.7).

Such a relation can be found only for 1D systems, or if we assume a 1D inhomogeneity of the observables. In higher dimensions our ability of constructing solvable quantum problems is limited by the possibility to analytically solve a classical hydrodynamics problem of reconstructing the density from the velocity or vice versa for an irrotational flow. Below we present a simple example of such a reconstruction.

6.3.1 Exact solution generated by a time-dependent scaling of observables

Let the evolution start from the ground state ψ_0 of a potential $V_0(\mathbf{r})$ with the ground state density $n_0(\mathbf{r})$ and the energy E_0. The simplest irrotational velocity field $\mathbf{v}(\mathbf{r}, t)$ for which Eq. (6.8) can be solved analytically is a linear function of coordinates with time-dependent coefficients

$$\mathbf{v}(\mathbf{r}, t) = \dot{\lambda}(t)[\mathbf{r} - \mathbf{r}_0(t)] + \dot{\mathbf{r}}_0(t). \tag{6.9}$$

This velocity corresponds to rigid motion of a fluid supplemented with a uniform expansion/compression with respect to the origin moving along the trajectory $\mathbf{r} = \mathbf{r}_0(t)$. The expansion/compression scaling factor is related to the parameter $\lambda(t)$, as $\alpha(t) = e^{\lambda(t)}$. This interpretation is confirmed by solving the continuity equation (6.8) with the velocity of Eq. (6.9). The corresponding solution for the density takes the form

$$n(\mathbf{r}, t) = \frac{1}{\alpha^3(t)} n_0 \left(\frac{\mathbf{r} - \mathbf{r}_0(t)}{\alpha(t)} \right), \tag{6.10}$$

which indeed corresponds to a rescaled density moving along the trajectory $\mathbf{r} = \mathbf{r}_0(t)$. The assumed initial conditions, $n(\mathbf{r}, t_0) = n_0(\mathbf{r})$ and $\mathbf{v}(\mathbf{r}, t_0) = 0$, are fulfilled if the time-dependent parameters λ and $\mathbf{r}_0$ have zero values and zero time derivatives at the initial time, that is $\lambda(t_0) = 0$, $\mathbf{r}_0(t_0) = 0$, $\dot{\lambda}(t_0) = 0$ and $\dot{\mathbf{r}}_0(t_0) = 0$.

Now we can insert the prescribed observables, Eqs. (6.9) and (6.10), into Eq. (6.7) to reconstruct the corresponding potential and the wave function.

To calculate the Bohm potential entering Eqs. (6.7a) we make use of the fact that $n_0(\mathbf{r})$ is the ground state density of the potential $V_0(\mathbf{r})$ with the energy E_0. This implies that the shifted and rescaled density $n(\mathbf{r}, t)$ of Eq. (6.10) corresponds to the instantaneous

ground state of the shifted and rescaled potential $\alpha^{-2}V_0\left((\mathbf{r}-\mathbf{r}_0)/\alpha\right)$ with the ground state energy $\alpha^{-2}E_0$. Therefore, the Bohm potential can be represented as

$$\frac{\nabla^2\sqrt{n(\mathbf{r},t)}}{2m\sqrt{n(\mathbf{r},t)}} = \frac{1}{\alpha^2(t)}V_0\left(\frac{\mathbf{r}-\mathbf{r}_0(t)}{\alpha(t)}\right) - \frac{1}{\alpha^2(t)}E_0. \tag{6.11}$$

The final results for the potential and the wave function generated by the velocity (6.9) or, equivalently by the density (6.10), take the following form

$$
\begin{aligned}
V(\mathbf{r},t) &= \frac{1}{\alpha^2}V_0\left(\frac{\mathbf{r}-\mathbf{r}_0}{\alpha}\right) - m\ddot{\mathbf{r}}_0\cdot\mathbf{r} \\
&\quad - \frac{m}{2}\frac{\ddot{\alpha}}{\alpha}(\mathbf{r}-\mathbf{r}_0)^2, \tag{6.12a} \\
\psi(\mathbf{r},t) &= \sqrt{\frac{1}{\alpha^3}n_0\left(\frac{\mathbf{r}-\mathbf{r}_0}{\alpha}\right)}\,e^{\mathrm{i}\varphi(\mathbf{r},t)}, \tag{6.12b} \\
\varphi(\mathbf{r},t) &= \frac{m}{2}\frac{\dot{\alpha}}{\alpha}(\mathbf{r}-\mathbf{r}_0)^2 + m\dot{\mathbf{r}}_0\cdot\mathbf{r} \\
&\quad - \int_0^t\left(\frac{1}{\alpha^2}E_0 + \frac{m}{2}\dot{\mathbf{r}}_0^2\right)dt' \tag{6.12c}
\end{aligned}
$$

Obviously, the first term in Eq. (6.12a) is the adiabatic potential. The other two terms describe two types of inertia forces – the usual linear acceleration force (the second term) and the inertial force related to a time-dependent deformation (the third term).

In the special case of a rigid motion, $\alpha = 1$ or $\lambda = 0$, only a linear acceleration inertial correction survives, so that the potential of Eq. (6.7a) simplifies to $V(\mathbf{r},t) = V_0(\mathbf{r}-\mathbf{r}_0) - m\ddot{\mathbf{r}}_0\cdot\mathbf{r}$. This potential rigidly transports a quantum system along a given trajectory without any reshaping of the initial density profile. It is worth noting that in this particular case our solution to the Schrödinger equation is not limited to one particle and can be trivially generalized to a system of any number of interacting identical particles. Indeed, the solution generated by a spatially uniform velocity field $\mathbf{v}(\mathbf{r},t) = \dot{\mathbf{r}}_0(t)$ can be obtained by the transformation to a uniformly accelerated reference frame [76]. Since the relative motion of particles is unaffected by this transformation the above potential will transport the centre of mass while keeping the quantum state for the relative motion unchanged. It is absolutely clear that if the initial state ψ_0 corresponds to that of the harmonic potential, our solution becomes identical to the harmonic potential theorem [75–77].

One can also easily see that the analytic solution of the Schrödinger equation for a harmonic oscillator with a time-dependent frequency $\omega(t)$ and a driving force $\mathbf{f}(t)$ [74]

$$i\partial_t \psi(\mathbf{r}, t) = \left(-\frac{\nabla^2}{2m} + \frac{1}{2}m\omega^2(t)\mathbf{r}^2 - \mathbf{f}(t) \cdot \mathbf{r}\right)\psi(\mathbf{r}, t), \tag{6.13}$$

is a particular case of our Eq. (6.12). Assuming $V_0(\mathbf{r}) = \frac{1}{2}m\omega_0^2\mathbf{r}^2$ in Eq. (6.12a) we find that the reconstructed potential $V(\mathbf{r}, t)$ coincides (up to irrelevant constant) with the potential in Eq. (6.13), where

$$\omega^2(t) = \frac{\omega_0^2}{\alpha^4} - \frac{\ddot{\alpha}}{\alpha}, \tag{6.14}$$

$$\mathbf{f}(t) = m\left(\frac{\omega_0^2}{\alpha^4} - \frac{\ddot{\alpha}}{\alpha}\right)\mathbf{r}_0 + m\ddot{\mathbf{r}}_0. \tag{6.15}$$

From these two equations we observe that the center of mass position $\mathbf{r}_0$ is the solution to the Newton equation for a driven harmonic oscillator

$$m\ddot{\mathbf{r}}_0 + m\omega^2(t)\mathbf{r}_0 = \mathbf{f}(t). \tag{6.16}$$

Hence in this particular case the solution of Schrödinger equation for the driven quantum oscillator is expressed in terms of the solution for a classical driven oscillator, which is the main observation made in Refs. [73, 74].

6.4   Inverse engineering of solvable quantum problems on a discrete space

In this section we describe and illustrate our general reconstruction strategy for lattice systems. In this case we use the maps for a generalized lattice-TDCDFT [105] to inverse engineer analytically solvable one-particle problems (or problems isomorphic to one-particle dynamics on a lattice) [105].

Our starting point is the Schrödinger equation for the wave function $\psi_i(t_0)$ describing a particle on an M-site lattice with time-dependent complex hopping parameters T_{ij},

$$i\partial_t \psi_i(t) = -\sum_{j=1}^{M} T_{ij}(t)\,\psi_j(t) \;\; ; \;\; T_{ii} = 0, \tag{6.17}$$

where indices i and j take values on the lattice sites indicating the position in the discrete space, and $T_{ij} = T_{ji}^*$ to have a Hermitian Hamiltonian. In the Schrödinger equation (6.17) we adopted a temporal gauge in which the scalar on-site potential and, possibly, a magnetic field enter via the phase of the hopping parameters $T_{ij}(t)$ [105]. For generality we also allow a time-dependent hopping rate $|T_{ij}(t)|$.

In the generalized lattice-TDCDFT of Ref. [105] the complex hopping $T_{ij}(t)$ plays the role of a driving potential. The corresponding observable in this approach can be called a "complex current" [105]

$$Q_{ij}(t) = 2T_{ij}(t)\psi_i^*(t)\psi_j(t). \tag{6.18}$$

The real part of $Q_{ij}(t)$ is equal to physical current J_{ij} on the lattice link connecting sites i and j, while its real part K_{ij} represents the kinetic energy on the link

$$Q_{ij} = K_{ij} + iJ_{ij}. \tag{6.19}$$

The link current $J_{ij}(t)$ and the on-site density $n_i(t) = |\psi_i(t)|^2$ are connected by the lattice continuity equation

$$\dot{n}_i(t) = -\sum_j J_{ij}(t). \tag{6.20}$$

Since the link current and the link kinetic energy are respectively antisymmetric and symmetric with respect to reversing the direction of the lattice link, $J_{ij} = -J_{ij}$ and $K_{ij} = K_{ij}$, the combined complex observable Q_{ij} is a Hermitian matrix, $Q_{ij} = Q_{ij}^*$.

Given the complex current $Q_{ij}(t)$ and the initial state $\psi_i(t_0) = |\psi_i(t_0)|e^{\varphi_i(t_0)} \neq 0$, the complex hopping $T_{ij}(t)$ and the wave function $\psi_i(t) = |\psi_i(t)|e^{\varphi_i(t)}$ can be expressed explicitly as functions of Q_{ij} and $\psi_i(t_0)$

$$T_{ij}(t) = \frac{Q_{ij}(t)}{2\psi_i^*(t)\psi_j(t)}, \tag{6.21a}$$

$$|\psi_i(t)| = \sqrt{|\psi_i(t_0)|^2 - \int_{t_0}^t \sum_j J_{ij}(t')\,\mathrm{d}t'}, \tag{6.21b}$$

$$\varphi_i(t) = \varphi_i(t_0) + \int_{t_0}^t \frac{\sum_j K_{ij}(t')}{2|\psi_i(t')|^2}\mathrm{d}t'. \tag{6.21c}$$

These formulas provide us with the analytic lattice-TDCDFT map from the observable to the conjugate driving potential and the corresponding solution of the time-dependent

Schrödinger equation . Using this map we can construct infinitely many analytically solvable problems generated by different time-dependent Hermitian observables $Q_{ij}(t)$.

Below we will give two examples which illustrate the possibility to analytically control quantum dynamics time in a discrete space.

6.4.1 Dynamics of one particle on a 1D chain

In this subsection we use our approach to manipulate the on-site density of a quantum particle on a finite tight-binding chain. Let us consider a particle on an atomic chain with a nearest neighbor hopping parameters $T_{i,i+1}$ of fixed amplitude $|T_{i,i+1}| = T_0$. The dynamics of the system is described by Eq. (6.17). Since for 1D systems only scalar (on-site) driving potentials are allowed, one can always gauge transform the Hamiltonian to the form with real hopping parameters $T_{i,i+1} = T_0$ and the real on-site potential $v_i(t)$.[105] In the new gauge, which is the lattice analog of the Coulomb gauge, the time-dependent Schrödinger equation reads:

$$i\partial_t \psi_i = -T_0(\psi_{i+1} + \psi_{i-1}) + v_i(t)\psi_i. \tag{6.22}$$

In the following, for definiteness, we assume that the evolution starts from the ground state ψ_0 of the chain.

The equations for the observables (6.18) and (6.19) for Q_{ij} , J_{ij} and K_{ij} remain the same except for the right hand side of Eq. (6.18) in which the hopping T_{ij} and the density matrix ρ_{ij} need to be replaced by their counterparts in the Coulomb gauge.

The map (6.21) from the complex current Q_{ij} to the hopping T_{ij} and the wave function ψ_i in the new gauge is transformed to an analytic map from Q_{ij} to the on-site potential v_i and the wave function ψ_i

$$v_i = \sum_{j=i}^{M} \Big(- \frac{K_{j,j+1} + K_{j-1,j}}{2n_j} + \frac{K_{j,j+1} + K_{j+1,j+2}}{2n_{j+1}}$$

$$+ \frac{\dot{J}_{j,j+1}K_{j,j+1} - J_{j,j+1}\dot{K}_{j,j+1}}{4T_0^2 n_i n_{j+1}} \Big), \tag{6.23a}$$

$$\psi_i(t) = \sqrt{n_i(t)}e^{i\varphi_i(t)}, \tag{6.23b}$$

$$\varphi_i(t) = \varphi_i(t_0) + \int_{t_0}^{t} \Big[\frac{K_{i,i+1} + K_{i-1,i}}{2n_i} - v_i\Big]dt'. \tag{6.23c}$$

The important point is that in the considered physical situation with the fixed hopping amplitude the above formulas are not sufficient to reconstruct the potential from the given dynamics of observables. The fixed value of the hopping amplitude sets an upper bound on allowed values of link currents. As a result not all possible Q become physically allowed, or v-representable in the TDDFT terminology. In fact, from the definition of Eq. (6.18) we find that the modulus of physically allowed $Q_{ij}(t)$ is bounded from above

$$|Q_{ij}(t)|^2 = 4T_0^2 n_i(t)n_j(t) < 4T_0^2. \tag{6.24}$$

Formally the condition of the fixed hopping amplitude reduces the dimension of the space $\mathcal{N}$ of observables. In the present case this restriction can be taken into account by expressing $J_{ij}(t)$ and $K_{ij}(t)$ in terms of on-site density $n_i(t)$. Firstly, in 1D we can solve the continuity equation (6.20) to get the link current

$$J_{i,i+1} = -\sum_{j=1}^{i} \dot{n}_j. \tag{6.25}$$

Secondly, we express $K_{ij}(t)$ in terms of $n_i(t)$ using Eqs. (6.24) and (6.25)

$$K_{i,i+1} = \pm\sqrt{4T_0^2 n_i n_{i+1} - \left(\sum_{j=1}^{i} \dot{n}_j\right)^2}, \tag{6.26}$$

where the sign is determined by the sign of $K_{ij}(t_0)$ at the initial time through the given initial state ψ_0.[106] Finally, by inserting Eqs. (6.25) and (6.26) into Eq. (6.23a) we obtain the explicit analytic formulas for the reconstruction of the on-site lattice potential and the corresponding wave function from a given time-dependent density distribution. These formulas correspond to the maps of the lattice TDDFT [103, 106]. It is interesting to note that exact solution proposed recently in Ref. [98] for a driven two-level system is, in fact, based on the above lattice TDDFT maps for a particular case of a two-site lattice.

Let us now demonstrate how this map works in practice by constructing a potential that produces prescribed evolution of a density. Consider a particle on an atomic chain with 11 sites and a positive hopping constant $T_0 = 1$ and assume that the dynamics starts

from the ground state of the chain with zero on-site potential,

$$\psi_i(0) = \psi_i^{gs} = \frac{1}{\sqrt{6}} \sin\left(\frac{\pi i}{12}\right), \tag{6.27}$$

We will construct the driving potential which generates the following two-stage evolution: (*i*) On the first stage for $0 < t < t_1$ the system evolves from the ground state of Eq. (6.27) to a state with a homogeneous density distribution $n_i = 1/11$; (*ii*) On the second stage for $t_1 < t < t_2$ the homogeneous density distribution shrinks to the center of the chain and by $t = t_2$ concentrates at site 6 with a Gaussian envelope, $n_i \propto e^{-(i-6)^2}$. The required time evolution of density $n_i(t)$ for this two-stage process is the following,

$$n_i(t) = \begin{cases} \frac{1}{11} S(t/t_1) + \left(1 - S(t/t_1)\right)|\psi_i^{gs}|^2 & 0 \leq t \leq t_1 \\ \\ \mathcal{N}(t) \exp\left[-S\left(\frac{t-t_1}{t_2-t_1}\right)(i-6)^2\right] & t_1 < t \leq t_2 \end{cases} \tag{6.28}$$

where $\mathcal{N}(t)$ is the normalization factor

$$\mathcal{N}(t) = \left(\sum_{i=1}^{11} e^{-S\left(\frac{t-t_1}{t_2-t_1}\right)(i-6)^2}\right)^{-1}. \tag{6.29}$$

Here $S(x)$ is a smooth step-like function which starts from zero at $x = 0$ and reaches unity at $x = 1$. For the reason that will be clear later we choose a function which has a zero first and second derivatives at $x = 0, 1$. Specifically here we use the following smooth step function which satisfies the above conditions

$$S(x) = x - \frac{1}{2\pi} \sin(2\pi x). \tag{6.30}$$

The time dependence of on-site densities $n_i(t)$ defined by Eq. (6.28) with $t_1 = 3$ and $t_2 = 12$ is shown on Fig. 6.2. Each line on this figure shows the prescribed evolution of the density on a particular site. At $t = 0$ the system is in the ground state (6.27), then it goes gradually to the homogeneous distribution at $t = 3$. Afterwards the density starts shrinking and finally at $t = 12$ it reaches a bell shaped Gaussian centered at the middle site.

The analytic representation for the corresponding driving potential now can be found immediately by inserting Eq. (6.28) into Eq. (6.23a) where the link current J_{ij} and the kinetic energy K_{ij} are given by Eqs. (6.25) and (6.26), respectively. Since for the
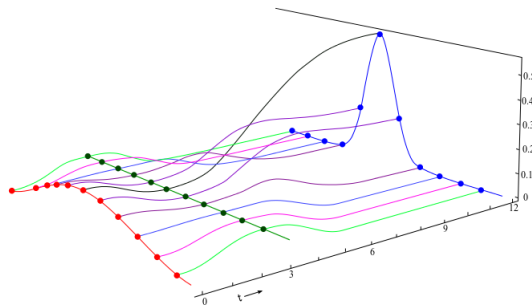
FIGURE 6.2: (Color online) The time evolution of the on-site densities determined by Eq. (6.28). Each line represents the dynamic of the density in a particular site. The time duration for the first stage is 3 units, $t_1 = 3$ and for the second stage is 9 units, $t_2 = 12$. Dots and their envelops indicate the density distribution in the initial $t = 0$, intermediate ($t = 3$), and the final ($t = 12$) states.

initial ground state $K_{ij}(0)$ is negative the minus sign must be chosen in Eq. (6.26), see section 3.4. Fig. 6.3 shows the on-site potentials for the first stage of the dynamics, $0 \le t \le 3$. Each curve represents the time dependence of the potential for a particular site. Similarly, Fig. 6.4 shows the driving potential for the second stage of the time evolution.
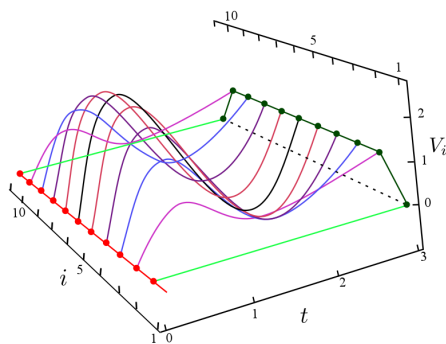
FIGURE 6.3: (Color online) On-site potentials (6.23a), for the first stage of the evolution, $0 \le t \le 3$, as a function of time. The on-site potentials for all sites are zero at the beginning. At $t = 3$ all v_i except those for the boundary sites reach 1, while the potentials for the two ending points stay zero. Color online.

Our reconstructed potential has one interesting property: on the first stage of the evolution the potential shown on Fig. 6.4 drives the system from its ground state at $t = 0$ an takes it into a state with a homogeneous density at $t = 3$ which is also the ground state of the system with the instantaneous potential $v_i(t = 3)$. Similarly, for the second stage of the evolution the driving potential shown in Fig. 6.4 takes the system from ground state and brings it into a new ground state with a Gaussian envelope. At the first glance this behavior looks surprising as the dynamics is by far non-adiabatic. The explanation
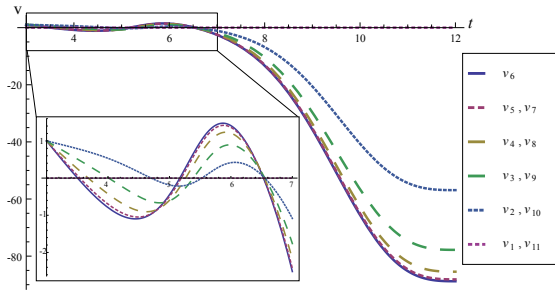
FIGURE 6.4: (Color online) On-site potentials (6.23a) as functions of time for the second stage of the evolution, $3 \leq t \leq 12$. The evolution for $3 < t < 7$, is zoomed in the magnified box.

is, however, simple. The system at $t = t_1$ and $t = t_2$ is in its instantaneous ground state because the first and second derivative of the step function S (6.30) are zero at those times and therefore the current J_{ij} and its first time derivative also vanish. By setting $J_{ij} = \dot{J}_{ij} = 0$ in Eq. (6.23a) we find the potential of the form

$$v_i = T_0 \sum_{j=i}^{M} \left(\frac{\sqrt{n_j} + \sqrt{n_{j+2}}}{\sqrt{n_{j+1}}} - \frac{\sqrt{n_{j-1}} + \sqrt{n_{j+1}}}{\sqrt{n_j}} \right). \tag{6.31}$$

This is a lattice analog of the Bohm potential that corresponds to the ground state potential for a given instantaneous density. Therefore our construction can be used to make a fast transfer of a system between different ground states.

6.4.2 Reconstruction of a driving magnetic field for a spin-1/2 system

In the last example we inverse engineer an analytically solvable Schrödinger equation for a driven spin-1/2 system, or equivalently, a generic two-level system. This formalism can be used, for example, to control the state evolution of a spin-1/2 using the time-dependent magnetic field. A similar problem has been addressed recently in Ref. [99]. Below we apply our general reconstruction strategy based on the lattice TDCDFT mapping.

Assume a spin in the initial state $|\psi_0\rangle$ subject to a time-dependent magnetic filed $\mathbf{B}(t)$. The time-dependent Schrödinger equation for the state vector $|\psi(t)\rangle$ reads

$$i\partial_t|\psi(t)\rangle = -\mathbf{B}(t) \cdot \hat{\mathbf{S}} \, |\psi(t)\rangle, \tag{6.32}$$

where $\hat{\mathbf{S}}$ is the spin-1/2 operator.

By a gauge transformation one can always eliminate z-component of the magnetic filed and therefore reduce the problem to solving the Schrödinger equation with the magnetic field in the xy-plane

$$i\partial_t \begin{pmatrix} \psi_\uparrow(t) \\ \psi_\downarrow(t) \end{pmatrix} = - \begin{pmatrix} 0 & \mathcal{B} \\ \mathcal{B}^* & 0 \end{pmatrix} \begin{pmatrix} \psi_\uparrow(t) \\ \psi_\downarrow(t) \end{pmatrix}, \tag{6.33}$$

where the "complex" magnetic field $\mathcal{B}$ is

$$\mathcal{B} = (B_x - iB_y)/2, \tag{6.34}$$

and $\psi_\uparrow(t) = \langle\psi(t)|\uparrow\rangle$ and $\psi_\downarrow(t) = \langle\psi(t)|\downarrow\rangle$ are the projections of the state vector on the eigenstates of $\hat{S}_z$.

Equation (6.33) is identical to the Schrödinger equation (6.17) for a two site lattice where the spin indecis $\uparrow$ and $\downarrow$ label the sites, and $\mathcal{B}(t)$ is the complex hopping parameter. Therefore we can directly apply the lattice-TDCDFT maps of Eqs. (6.21) to reconstruct the driving magnetic field and the wave functions $\psi_{\uparrow,\downarrow} = |\psi_{\uparrow,\downarrow}|e^{i\varphi_{\uparrow,\downarrow}}$ from given dynamics of the complex observable $Q(t) = K(t) + iJ(t)$ defined in Eq. (6.18). In the present case the reconstruction formulas reduce to the form

$$|\psi_{\uparrow,\downarrow}(t)| = \sqrt{|\psi_{\uparrow,\downarrow}(0)|^2 \mp \int_0^t \mathrm{Im}Q(t')\,\mathrm{d}t'}, \tag{6.35a}$$

$$\varphi_{\uparrow,\downarrow}(t) = \varphi_{\uparrow,\downarrow}(0) + \int_0^t \frac{\mathrm{Re}Q(t')}{2|\psi_{\uparrow,\downarrow}(t')|^2}\mathrm{d}t', \tag{6.35b}$$

$$\mathcal{B}(t) = \frac{Q(t)}{2\psi_\uparrow^*(t)\psi_\downarrow(t)}. \tag{6.35c}$$

Equations (6.35) provide an analytic parametrization of the driving field and the wave function in terms of a given trajectory in the two-dimensional space $\mathcal{N} = \{K, J\}$ of observables. The point in the space $\mathcal{N}$ corresponds to a given kinetic energy K and intersite current J for a particle on the two-site lattice. This physical parametrization is universally applicable to lattice systems with any number of sites. In the particular two-site case one can propose an alternative parameterization of the driving field, which has an intuitive interpretation in the physical context of the spin-1/2 system. Below we map the space $\mathcal{N} = \{K, J\}$ onto a Bloch sphere and rearrange Eqs. (6.35) accordingly to relate the driving field to a given trajectory in the projective Hilbert space for spin-1/2.

As the first step we represent the state vector of spin-1/2 as follows

$$|\psi(t)\rangle = e^{i\beta}\left[\cos(\theta/2)|\uparrow\rangle + e^{i\phi}\sin(\theta/2)|\downarrow\rangle\right] \tag{6.36}$$

where $\theta(t)$ and $\phi(t)$ are the spherical angles representing a point on the Bloch sphere, and $\beta(t)$ is an overall phase of the wave function. Next, to map the trajectory $Q(t)$ to a trajectory on the Bloch we use Eq. (6.35) and express Q in terms of the wave function amplitudes $|\psi_{\uparrow,\downarrow}|$ and the relative phase ϕ

$$Q = 2\dot{\phi}\frac{|\psi_\uparrow|^2|\psi_\downarrow|^2}{|\psi_\uparrow|^2 - |\psi_\downarrow|^2} - i\partial_t|\psi_\uparrow|^2. \tag{6.37}$$

By substituting $|\psi_{\uparrow,\downarrow}|$ from Eq. (6.36) we relate the complex coordinate Q in the space $\mathcal{N}$ to the spherical coordinates (θ, ϕ) on the Bloch sphere

$$Q = \frac{1}{2}\left(\dot{\phi}\sin\theta\tan\theta + i\,\dot{\theta}\sin\theta\right). \tag{6.38}$$

This equation gives the required map between the trajectory in the original space of observables to the corresponding trajectory of spin-1/2 on the Bloch sphere. Finally, by inserting Q of Eq. (6.38) into Eq. (6.35c) for $\mathcal{B}$ (6.35c) we get a new analytic representation for components of the magnetic field

$$B_x = \dot{\phi}\tan\theta\cos\phi + \dot{\theta}\sin\phi, \tag{6.39a}$$

$$B_y = -\dot{\theta}\cos\phi + \dot{\phi}\tan\theta\sin\phi. \tag{6.39b}$$

The spherical coordinates (θ, ϕ) determine the wave function Eq. (6.36) up to a common phase β. The phase $\beta = \varphi_\uparrow$ is calculated directly from Eq. (6.35b) by substituting the expressions of $|\psi_\uparrow|$ and ReQ in terms of θ and ϕ,

$$\beta(t) = \beta(0) + \frac{1}{2}\int_0^t dt'\dot{\phi}\tan\theta\tan\frac{\theta}{2}. \tag{6.40}$$

Equations (6.39), (6.36), and (6.40) solve the problem of reconstructing the driving field and the wave function from a given trajectory on the Bloch sphere.

Equations (6.39) demonstrate one subtlety, which is very similar to the v-representability problem in TD(C)TDFT. Not all trajectories on the Bloch sphere are physically reproducible if the driving magnetic field is limited to the (x, y)-plane. For example, it is

impossible to drive the system along the equator $(\pi/2, \phi(t))$ with a finite magnetic field because the right hand side in Eq. (6.39) diverges at $\theta = \pi/2$. Similarly, any trajectory which causes a divergence in right hand side of Eq. (6.39) is not $\mathcal{B}$-representable. All physically allowed trajectories when crossing the equator should approach it in a way that $\dot{\phi} \tan \theta$ stays finite, which translates to the condition $\dot{\phi} \to 0$ when $\theta \to \pi/2$. In other words, a physical trajectory, generated on the Bloch sphere by an in-plane magnetic field, can cross the line $\theta = \pi/2$ only if it is perpendicular to that line at the crossing point. This is absolutely clear physically because at any instant the magnetic filed generates rotation of the spin vector about the direction of $\mathbf{B}$. Therefore the initially coplanar to $\mathbf{B}$ spin is always driven out of the plane. Apparently when reconstructing the driving field from a trajectory on the Bloch sphere we should take it from a $\mathcal{B}$-representable set containing trajectories which either do not touch the equator or cross it perpendicularly.

Now we are ready to present an explicit example of the reconstruction.

Analytically controlled spin flip: design of a quantum NOT gate

To illustrate our inverse engineering formulas we construct a control pulse which does the operation. Initially the system is in the ground state corresponding to some magnetic field $\mathbf{B}(0)$. During the pulse duration τ the magnetic field is changing and at the end of the pulse returns to its initial value $\mathbf{B}(\tau) = \mathbf{B}(0)$ while the system is driven to the excited state in the field $\mathbf{B}(0)$. Therefore after the pulse the Hamiltonian returns to the initial form, but the direction of the spin is reversed.

For definiteness we assume the initial/final field in the x-direction, $\mathbf{B}(0) = \mathbf{B}(\tau) = \hat{\mathbf{x}} B_0$. Therefore the initial state is $| \leftarrow \rangle = 1/\sqrt{2}(| \uparrow \rangle + | \downarrow \rangle)$. The target state which should be reached at the end of the pulse corresponds to another eigenstate of $\hat{S}_x$, that is $| \rightarrow \rangle = 1/\sqrt{2}(| \uparrow \rangle - | \downarrow \rangle)$. On the Bloch sphere the initial and the final (target) states correspond to the points $(\pi/2, 0)$ and $(\pi/2, \pi)$, respectively.

The first step in constructing the required control pulse is to find a $\mathcal{B}$-representable trajectory which at $t = 0$ starts at the point $(\pi/2, 0)$ and arrives to the point $(\pi/2, \pi)$ at the time $t = \tau$. We note either boundary point belongs to the equatorial line. Therefore the trajectory should leave the initial point and arrive to the final point along the corresponding meridians. In order take care of the $\mathcal{B}$-representability automatically we

introduce a new independent variable $\gamma = \dot{\phi}\tan\theta$. The angle θ in the relevant range of $0 < \theta < \pi$ is related to the new variable as follows

$$\theta = 2\arctan\left[\sqrt{(\dot{\phi}/\gamma)^2 + 1} - \dot{\phi}/\gamma\right]. \tag{6.41}$$

Now the "dangerous" equatorial points correspond to the points of the trajectory with $\dot{\phi}/\gamma = 0$.

By re-expressing the magnetic field of Eqs. (6.39) and the common phase β of Eq. (6.40) in terms of γ and ϕ we find

$$B_x = \gamma\cos\phi - \frac{\partial_t(\dot{\phi}/\gamma)}{(\dot{\phi}/\gamma)^2 + 1}\sin\phi, \tag{6.42a}$$

$$B_y = \frac{\partial_t(\dot{\phi}/\gamma)}{(\dot{\phi}/\gamma)^2 + 1}\cos\phi + \gamma\sin\phi. \tag{6.42b}$$

and

$$\beta(t) = \beta(0) + \frac{1}{2}\int_0^t dt'\left[\gamma\sqrt{(\dot{\phi}/\gamma)^2 + 1} - \dot{\phi}\right]. \tag{6.43}$$

Now we need to find two functions $\gamma(t)$ and $\phi(t)$ which will do the required job. The first obvious set of conditions for $\phi(t)$ is

$$\phi(0) = 0, \quad \phi(\tau) = \pi, \quad \text{and} \quad \dot{\phi}(0) = \dot{\phi}(\tau) = 0. \tag{6.44}$$

It follows from Eq. (6.42) that the requirement $\mathbf{B}(0) = \mathbf{B}(\tau) = B_0\hat{\mathbf{x}}$ will be fulfilled if

$$\gamma(0) = -\gamma(\tau) = B_0, \tag{6.45}$$

and $\partial_t(\dot{\phi}/\gamma) = 0$ at the boundary points, $t = 0, \tau$. The latter condition is satisfied if at $t = 0, \tau$ the second derivative of $\phi(t)$ vanishes

$$\ddot{\phi}(0) = \ddot{\phi}(\tau) = 0. \tag{6.46}$$

In addition we have to make sure that the ratio $\dot{\phi}/\gamma$ is finite for all $0 < t < \tau$.
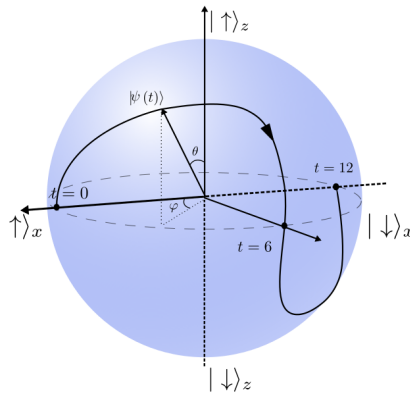
FIGURE 6.5: Trajectory of the spin state on the Bloch sphere generated by Eqs. (6.47) for $\tau = 12$ and $B_0 = 1$. The trajectory leaves ground state $| \leftarrow \rangle$ and arrives to the excited state $| \rightarrow \rangle$ perpendicularly ($\dot{\phi} = 0$) to the equatorial line.

As an example we suggest the following $\gamma(t)$ and $\phi(t)$ which fulfill all above conditions

$$\gamma(t) = \frac{1}{2} B_{x0}(1 - 2t/\tau)\big[(1 - 2t/\tau)^2 + 3\big], \tag{6.47a}$$

$$\varphi(t) = \frac{\pi t}{\tau} - \frac{1}{4}\sin(\frac{4\pi t}{\tau}). \tag{6.47b}$$

Here $\gamma(t)$ is a smooth monotonically decreasing function antisymmetric with respect to the point $t = \tau/2$. It goes from $-B_0$ to B_0 and crosses zero at the middle of the pulse. As an extra condition we require that $\dot{\gamma}(0) = \dot{\gamma}(\tau) = 0$ which allows to smoothly continue the driving field beyond the interval $0 < t < \tau$. The function $\phi(t)$ in Eq. (6.47b) increases monotonically from 0 to π and has zero first and second derivatives at the boundary points $t = 0$ and $t = \tau$, and at $t = \tau/2$. The derivative $\dot{\phi}(t) = \frac{2\pi t}{\tau}\sin^2(\frac{4\pi t}{\tau})$ is symmetric with respect to the middle point $t = \tau/2$.

The corresponding trajectory on the Bloch-sphere is shown in Fig. 6.5 for $B_0 = 1$ and $\tau = 12$. The trajectory starts from the state $| \leftarrow \rangle$ on the equator, goes to the upper hemisphere, then at $t = \tau/2$ it crosses the equatorial line at the point $(\pi/2, \pi/2)$ and reaches the final state $| \rightarrow \rangle$ from the lower hemisphere. Because of the special symmetry of the generating functions the trajectory has a central symmetry with respect to the middle point.

Substituting $\gamma(t)$ and $\phi(t)$ into Eqs. (6.42) we find magnetic field generating this dynamics. In Fig. 6.6 we plot the path of the time-dependent magnetic field vector in the xy-plane. Each dot represents the magnetic field vector at integer times from 0 to 12.
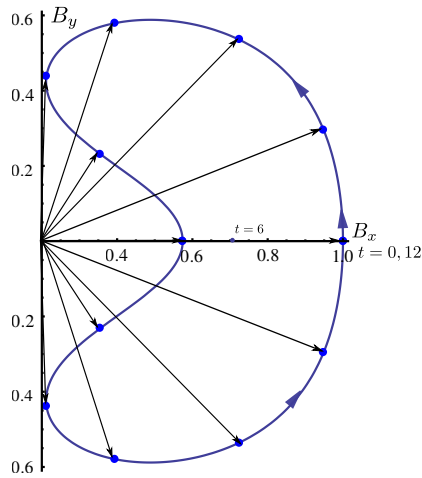
FIGURE 6.6: The trajectory traced out the magnetic field vector (6.42) in the xy-plane. Starting from the initial value $\mathbf{B}(0) = \hat{\mathbf{x}}$ the magnetic field follows the trajectory clockwise and comes back at $t = \tau = 12$. Each arrow represents the magnetic field vector at integer times $t = 0, 1, \ldots 12$.

The corresponding wave function is given by Eq. (6.36) with the common phase $\beta(t)$ defined after Eq. (6.43). For our particular example we easily find the overall phase at the end of the pulse, $\beta(\tau) = \beta(0) - \pi/2$. [2] Hence by the end of the pulse, the system which initially was in the ground state is transferred to the excited state and the wave function acquires a common phase shift of $\pi/2$.

One can check that if the pulse defined by Eqs. (6.42) and (6.47) is applied to the initial excited state $|\psi(0)\rangle = |\rightarrow\rangle$, the system is driven to the ground state $|\leftarrow\rangle$. Therefore our pulse can be viewed as a realization of a quantum NOT gate which transforms (up to a common phase shift) $|\leftarrow\rangle$ into $|\rightarrow\rangle$, and $|\rightarrow\rangle$ into $|\leftarrow\rangle$.

---

[2]Because $\gamma(t)$ is antisymmetric and $\dot{\phi}(t)$ is symmetric with respect to $\tau/2$, the integral of the term with the square root vanishes. Therefore we are left with the result $\beta(\tau) = \beta(0) - [\phi(\tau) - \phi(0)]/2$.

Chapter 7

Dicussion and Conclusion

In this dissertation we studied the very core principles of the TDDFT and then extended it beyond its standard formalism and applications. Following the flow of the thesis we start the conclusion by an ideological comparison between DFT and TDDFT.

DFT as a theory based on the static quantum mechanics focuses on the single state with minimal energy.[2] As a result the DFT formulation is a direct consequence of the minimum principle which allows for a variational search of the universal ground state functional. In contrast, in quantum dynamics we are dealing with the initial value problem. Since there is no minimum principle in dynamics, except in a few special cases [128–130], a direct extension of DFT is not possible. One of the breakthroughs of the Runge-Gross work was their approach to the problem by explicit use of the equations of motion. This led to the first sufficiently generic proof of the unique time-dependent density-potential mapping which pioneered a major advancement in the study of quantum dynamics. The Runge-Gross proof of the uniqueness using the power series expansion is reminiscent to the Taylor series-based approach by Cauchy to the theory of ODE. Despite a relatively straightforward proof of uniqueness the proof for the existence is much more involved, as one needs to prove the convergence of the Taylor series.[131] With this comparison it should not be very surprising that the v-representability problem has not been fully solved yet.

However there is an alternative approach to the density-potential map, inspired by the Runge-Gross theorem, in which the TDDFT map is formulated as a non-linear many-body problem (NLSE).[9, 13–15] This approach led to the three most general proofs of

v-representability and uniqueness which include the fixed-point proof of TDDFT under the assumption of boundness for a certain response function [17] , the rigorous proof of TDCDFT on a lattice [16] and the rigorous lattice-TDDFT proof in chapter 3.

The NLSE formulation, as a well posed mathematical problem, opens the possibility to study the density-potential map using the established theorems for non-linear PDEs.[114, 115] Realization of this program requires a solution of two mathematical problems. The first one, solved recently, is to prove the invertibility of the Sturm-Liouville operator in the force balance equation. [112, 113] The second one, still open, is to show the Lipschitz continuity of the inverted Sturm-Liouville operator in the proper functional space.

The complexity of working with infinite-dimensional functional spaces motivated us to approach the problem from a different direction and to prove the basic mapping for a lattice-regularized quantum system with a finite dimensional Hilbert space. This is relevant since truncating the Hilbert space is an inevitable approximation in modeling physical systems on the computer. Formally it reduces a non-linear PDE to a set of ODEs while the force balance equation becomes purely algebraic. As a result of such reduction we were able to prove the basic existence and uniqueness theorem using standard results from the theory of nonlinear ODE (see chapter 3). As we expected from the previous works [20–22, 24, 103] the lattice TDDFT is not unconditionally valid. Similarly to the lattice TDCDFT, Ref. [16], apart from a physical restriction on the time dependence of the density, there are also limitations on possible many-body initial states, which should be in Ω, the v-representable subset of the Hilbert space. We proved that this condition is fulfilled for any ground state of a connected lattice and argued that most physical states should belong to Ω

The proof of the basic theorem for the lattice TDDFT presented in this thesis as well as the proof for the lattice TDCDFT [16] essentially rely on the concept of the v-representability subset Ω in the Hilbert space. It would be very interesting and useful for the future to carefully study and characterize the structure/geometry of this set. Formally the v-representable subset is defined as a part of the Hilbert space where the lattice analogue of the Sturm-Liouville operator has a nonzero determinant. Similarly to the explicit example of Section 3.4, in general the v-representability subset Ω should consist of two regions with different signs of the determinant which are separated by a surface. What is the geometry of each part for a general quantum system? Are

they simply- or multiply-connected? Is it possible for a system driven by a physical potential to cross the surface or is it forever confined to one initially fixed subregion of Ω? Answering these questions will definitely deepen our understanding of TDDFT-related theories as well as quantum dynamics in general. In chapter 5, by considering a special physical example, we found that the answer to the second question is "yes". In the dynamics like Rabi oscillations for a single particle system the state population is completely transferred to the excited state and therefore the v-representability boundary is crossed.

All mathematically rigorous proofs presented in this work are limited to lattice systems. However taking the continuum limit accurately is a highly nontrivial but very interesting problem. A careful generalization of the lattice TDDFT to the continuum limit should answer a fundamental question of whether or not there is a counterpart of the v-representability subset Ω in the continuum TDDFT.

The rigorous formulation of TDDFT for lattice electrons driven by the classical external field suggests a possibility to go beyond the standard setup and take into account the quantum nature of the driving electromagnetic field. For this system as a result of added photonic degree of freedom to the wave function we expect that the minimal set of the basic variables fully describing the state of the system must include an extra observable. The extra observable, as suggested in the Taylor expansion-based proof of QED-TDDF Ref. [40], is the expectation value of the photonic canonical coordinate. Therefore the basic map in QED-TDDFT is from the particle density and the field average to the external driving potentials. The latter is a scalar potential driving the electronic sector and a time dependent dipole moment driving the photonic coordinate.

Again the v-representability and uniqueness is equivalent to the existence of a unique solution to a certain system of NLSE. The main difference and, in fact, the main mathematical difficulty of QED-TDDFT , compared to the purely electronic lattice TDDFT is the existence of continuum variables describing photonic modes. As a result the Hilbert space becomes an infinite dimensional functional space, and the universal NLSE turns into a system of PDEs. This puts the lattice QED-TDDFT, in a certain sense, on half way between the electronic lattice TDDFT and the usual TDDFT in the continuum space. Using the theory of PDE we showed the corresponding Cauchy problem indeed has a unique solution . Therefore, the unique QED-TDDFT map exists, provided, the

basic variables have continuous second time-derivatives and the initial state belongs to some well defined v-representability subset of the Hilbert space Ω. We further extended the ground state v-representability theorem to a very general electron-photon lattice Hamiltonian. If a system evolves from the ground state of such a Hamiltonian the local v-representability is always guaranteed. The simplest non-trivial example of this construction is a system of a one-electron Hubbard dimer coupled to a single photonic mode, which is identical to the Rabi and spin-boson models.[42–44]

Apparently any practical application for QED-TDDFT requires development of approximations to the universal exchange-correlation functional. Despite the fact that this theory is still very young there is an active research in developing such approximate functionals. [132].

In the second half of the thesis we focused on practical aspects of TDDFT. In chapter 5 we performed a qualitative and quantitative study of the non-adiabatic features in the TDDFT functional for a Hubbard dimer to exemplify the performance of the most widely used adiabatic approximation. We chose this system for two main reasons: First, its Kohn-Sham functional is analytic and known and second, the exact ground state adiabatic exchange-correlation functional can be calculated using the Levy-Lieb constrained search. [66, 110, 111]

We propagated the Kohn-Sham equation self-consistently by using the adiabatic exchange-correlation functional and found that the non-adiabaticity is crucial to properly capture the physics of resonant and slightly detuned Rabi oscillations. In section 5.4 we used the analytic density potential map to derive an explicit fully non-adiabatic Hartree-exchange-correlation functional that accurately describes resonant and slightly detuned Rabi oscillations. Moreover this quasi-exact functional incorporates explicitly the initial-state dependence and is non-local in time.

In addition, this functional highlights another important feature of the lattice TDDFT – the dependence of the exact exchange-correlation functional on the second time-derivative of the density. This dependence has been proven controversial and even raised questions about usability of the true exchange-correlation potential.[133, 134] Essentially the point is that one cannot propagate the self-consistent Kohn-Sham equation if the potential depends on the second time-derivative of the density. Calculation of the

second derivative at a given time t requires knowledge of the future density. This seems to prevent using the exact exchange-correlation potential in an iterative scheme.

Fortunately, this does not create any problem because lattice TDDFT, and most likely standard TDDFT, holds true under the assumption of continuous second time-derivative of the density. Therefore $\ddot{n}$ can be found by differentiating the first time-derivative from the left. The only problem is with the density at the initial time which has no defined history. However, we can determine the second derivative of the density through the interacting force balance equation for a given interacting initial state and external potential at t_0. Therefore, for the Kohn-Sham propagation all we need apart from the exchange-correlation functional are the external potential, the interacting initial states, and the Kohn-Sham initial state.

In chapter 6 we continued our work on practical applications by proposing a new use for the analytic maps in TD(C)DFT. In this approach we parametrized both the wave function and the driving potential in terms of intuitive physical observables, e.g. the density and/or the current, which allows us to design analytical control signals for pre-scribed dynamics. With this strategy we are able to inverse engineer infinitely many analytically solvable time-dependent problems covering dynamics both in a real space and on lattices. Moreover, for problems like the state preparation in which the initial and target states are known it allows us to incorporate some favourable features, such as minimal initialization time into the time-evolution.

We considered a number of situations in which the TD(C)DFT maps are known analytically. In all those cases the basic observables, such as the density or the current, were used for a convenient and physically intuitive parametrization of the time-evolution. As the first pedagogical example we considered the control problem for the Schrödinger equation to describe dynamics of a single quantum particle driven by a time-dependent electromagnetic field. If the dynamics is restricted to be generated by an electric field, the solution can be reconstructed from the dynamics of the density or, equivalently, from the given (irrotational) velocity field. From the general reconstruction formulas one can recover the known analytic solutions of the time-dependent Schrödinger equation for a harmonic oscillator driven by a time-dependent frequency [73, 74].

Afterwards we applied our strategy to a problem of quantum dynamics on lattices

(discrete spaces). We used the analytic maps for the one-particle generalized lattice-TDCDFT [105] where the basic observables are the intersite current and the kinetic energy. As the first illustration of this setup we considered a manipulation of a one particle state on a finite tight-binding chain. We constructed an analytic driving field that generates a fast reshaping of the initial ground state density to the ground state density of another potential. In the second example we demonstrated the engineering of analytically solvable two level Schrödinger equations describing, in particular, dynamics of spin-1/2 system driven by a time-dependent magnetic field. We constructed a cyclic analytic control pulse which works as a quantum NOT gate, that is, it flips the direction of the spin for two basis states.

This universal strategy further develops and extends promising applications of TDDFT to the quantum control.[107, 108] More importantly, it connects the ideas of the density-potential mapping in TDDFT and TDCDFT to a wider range of coherent control [135] and the state preparation problems in the quantum computing [81–83].

To summarize, we hope this thesis will contribute to a better understanding of quantum dynamics driven by external time-dependent potentials. We mainly focused on quantum dynamics on a lattice for two very important reasons. First of all, it is extremely popular in computational physics and, second, it simplifies the mathematical construction of the problem. This strategy led to number of rigorous theorems for TDDFT and QED-TDDFT which provide a firm foundation for most practical applications in these formalisms. However, there are still interesting questions to be addressed in the future. For example, we do not fully understand all the physical situations when the boundary of v-representability subset is reached. In the study of non-linear dynamics we quantified the importance of the history and initial value dependence in exchange-correlation functional. Finally, we proposed a new application of the TD(C)DFT maps as we introduced them to new fields such as quantum control and quantum computing. We believe that with the development of accurate approximations to the exchange-correlation functional a similar strategy can be applied to many-body dynamics.

Appendix A

Derivation of the equations of motions

A.1 Continuity equation on a lattice

To derive the continuity equation on a lattice we first start from the second quantized version of the Hamiltonian for a system of N electrons on an M-site lattice with Schrödinger equation (3.1)

$$
\begin{aligned}
\hat{H} &= \hat{T} + \hat{V}_{ext} + \hat{W} \\
&= \sum_{\mathbf{r},\mathbf{r}'} T_{\mathbf{r},\mathbf{r}'} \hat{a}_{\mathbf{r}}^{\dagger} \hat{a}_{\mathbf{r}'} + \sum_{\mathbf{r}} v_{\mathbf{r}} \hat{a}_{\mathbf{r}}^{\dagger} \hat{a}_{\mathbf{r}} + \frac{1}{2} \sum_{\mathbf{r},\mathbf{r}'}^{M} w_{\mathbf{r},\mathbf{r}'} \hat{a}_{\mathbf{r}}^{\dagger} \hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}} \hat{a}_{\mathbf{r}'},
\end{aligned}
\tag{A.1}
$$

where $\hat{a}_{\mathbf{r}}^{\dagger}$ and $\hat{a}_{\mathbf{r}}$ are the electronic creation and annihilation operators at site $\mathbf{r}$ respectively. The summation is over the lattice site coordinates $\mathbf{r}$.

The creation and annihilation operators obey the following anti-commutation relations

$$
\{\hat{a}_{\mathbf{r}}, \hat{a}_{\mathbf{r}'}^{\dagger}\} = \delta_{\mathbf{r},\mathbf{r}'},
\tag{A.2}
$$

$$
\{\hat{a}_{\mathbf{r}}, \hat{a}_{\mathbf{r}'}\} = \{\hat{a}_{\mathbf{r}}^{\dagger}, \hat{a}_{\mathbf{r}'}^{\dagger}\} = 0.
\tag{A.3}
$$

The on-site density operator in the second quantization reads:

$$
\hat{n}(\mathbf{r}) = \hat{a}_{\mathbf{r}}^{\dagger} \hat{a}_{\mathbf{r}}.
\tag{A.4}
$$

One can calculate the time derivative of the on-site density using the Heisenberg equation

$$\frac{d}{dt}\hat{n}(\mathbf{r}) = i[\hat{H}, \hat{n}(\mathbf{r})]. \tag{A.5}$$

The only non-commuting term in the Hamiltonian (A.1) is the kinetic term. [1] So we have [2]

$$
\begin{aligned}
[\hat{T}, \hat{n}(\mathbf{r})] &= \sum_{\mathbf{r}',\mathbf{r}''} T_{\mathbf{r}',\mathbf{r}''} [\hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}''}, \hat{a}_{\mathbf{r}}^{\dagger} \hat{a}_{\mathbf{r}}] \\
&= \sum_{\mathbf{r}',\mathbf{r}''} T_{\mathbf{r}',\mathbf{r}''} \left(\hat{a}_{\mathbf{r}'}^{\dagger} [\hat{a}_{\mathbf{r}''}, \hat{a}_{\mathbf{r}}^{\dagger} \hat{a}_{\mathbf{r}}] + [\hat{a}_{\mathbf{r}'}^{\dagger}, \hat{a}_{\mathbf{r}}^{\dagger} \hat{a}_{\mathbf{r}}] \hat{a}_{\mathbf{r}''} \right) \\
&= \sum_{\mathbf{r}',\mathbf{r}''} T_{\mathbf{r}',\mathbf{r}''} \left(\hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}} \{\hat{a}_{\mathbf{r}''}, \hat{a}_{\mathbf{r}}^{\dagger}\} - \hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}}^{\dagger} \{\hat{a}_{\mathbf{r}''}, \hat{a}_{\mathbf{r}}\} + \{\hat{a}_{\mathbf{r}'}^{\dagger}, \hat{a}_{\mathbf{r}}^{\dagger}\} \hat{a}_{\mathbf{r}} \hat{a}_{\mathbf{r}''} - \{\hat{a}_{\mathbf{r}'}^{\dagger}, \hat{a}_{\mathbf{r}}\} \hat{a}_{\mathbf{r}}^{\dagger} \hat{a}_{\mathbf{r}''} \right),
\end{aligned}
$$

Using the anti-commutation relations (A.2).

$$[\hat{T}, \hat{n}(\mathbf{r})] = \sum_{\mathbf{r}'} T_{\mathbf{r}',\mathbf{r}} \left(\hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}} - \hat{a}_{\mathbf{r}}^{\dagger} \hat{a}_{\mathbf{r}'} \right). \tag{A.6}$$

There is no external magnetic field so $T_{\mathbf{r},\mathbf{r}'} = T_{\mathbf{r}',\mathbf{r}}$. In the case of magnetic field the indeces are non-interchangeable so we would have: $\left(T_{\mathbf{r}',\mathbf{r}} \hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}} - T_{\mathbf{r},\mathbf{r}'} \hat{a}_{\mathbf{r}}^{\dagger} \hat{a}_{\mathbf{r}'} \right)$

Substituting back into Eq. (A.7) we have the continuity equation in the operator form

$$\frac{d}{dt}\hat{n}(\mathbf{r}) = i \sum_{\mathbf{r}'} T_{\mathbf{r}',\mathbf{r}} \left(\hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}} - \hat{a}_{\mathbf{r}}^{\dagger} \hat{a}_{\mathbf{r}'} \right). \tag{A.7}$$

taking the expectation value with respect to $\psi(\mathbf{r}_1, ..., \mathbf{r}_N; t)$ we have

$$\dot{n}(\mathbf{r}; t) = \sum_{\mathbf{r}'} T_{\mathbf{r}',\mathbf{r}} \left(\rho(\mathbf{r}', \mathbf{r}) - \rho(\mathbf{r}, \mathbf{r}') \right), \tag{A.8}$$

where

$$\rho(\mathbf{r}', \mathbf{r}) = N \sum_{\mathbf{r}_2, ..., \mathbf{r}_N} \psi^*(\mathbf{r}', \mathbf{r}_2, ..., \mathbf{r}_N) \psi^*(\mathbf{r}, \mathbf{r}_2, ..., \mathbf{r}_N). \tag{A.9}$$

---

[1] $[\hat{A}, f(\hat{A})] = 0.$
[2] $[\hat{A}\hat{B}, \hat{C}] = \hat{A}[\hat{B}, \hat{C}] + [\hat{A}, \hat{C}]\hat{B}, \quad [\hat{A}\hat{B}, \hat{C}] = \hat{A}\{\hat{B}, \hat{C}\} - \{\hat{A}, \hat{C}\}\hat{B}$

A.2 Force balance equation on a lattice

To derive the force balance equation (3.8) we need to calculate the second time derivative of the density which means to calculate

$$\frac{d^2}{dt^2}\hat{n}(\mathbf{r}) = i[\hat{H}, \dot{\hat{n}}(\mathbf{r})]. \tag{A.10}$$

It is an straight forward but lengthy job to calculate the commutator in the right hand side of this equation. First we substitute the derivative of the density from the operator form of the continuity equation (A.7)

$$
\begin{aligned}
[\hat{H}, \dot{\hat{n}}(\mathbf{r})] &= i\sum_{\mathbf{r}'} T_{\mathbf{r}',\mathbf{r}}[\hat{H}, \hat{a}^\dagger_{\mathbf{r}'}\hat{a}_{\mathbf{r}} - \hat{a}^\dagger_{\mathbf{r}}\hat{a}_{\mathbf{r}'}] \\
&= i\sum_{\mathbf{r}'} T_{\mathbf{r}',\mathbf{r}}\left([\hat{T}, \hat{a}^\dagger_{\mathbf{r}'}\hat{a}_{\mathbf{r}} - \hat{a}^\dagger_{\mathbf{r}}\hat{a}_{\mathbf{r}'}] + [\hat{V}_{ext}, \hat{a}^\dagger_{\mathbf{r}'}\hat{a}_{\mathbf{r}} - \hat{a}^\dagger_{\mathbf{r}}\hat{a}_{\mathbf{r}'}] + [\hat{W}, \hat{a}^\dagger_{\mathbf{r}'}\hat{a}_{\mathbf{r}} - \hat{a}^\dagger_{\mathbf{r}}\hat{a}_{\mathbf{r}'}]\right).
\end{aligned}
\tag{A.11}
$$

We calculate each commutator separately and then we will put them all together

$$
\begin{aligned}
[\hat{T}, \hat{a}^\dagger_{\mathbf{r}'}\hat{a}_{\mathbf{r}}] &= \sum_{\mathbf{r}'',\mathbf{r}'''} T_{\mathbf{r}'',\mathbf{r}'''}([\hat{a}^\dagger_{\mathbf{r}''}\hat{a}_{\mathbf{r}'''}, \hat{a}^\dagger_{\mathbf{r}'}\hat{a}_{\mathbf{r}}] \\
&= \sum_{\mathbf{r}'',\mathbf{r}'''} T_{\mathbf{r}'',\mathbf{r}'''}\left(\hat{a}^\dagger_{\mathbf{r}''}[\hat{a}_{\mathbf{r}'''}, \hat{a}^\dagger_{\mathbf{r}'}\hat{a}_{\mathbf{r}}] + [\hat{a}^\dagger_{\mathbf{r}''}, \hat{a}^\dagger_{\mathbf{r}'}\hat{a}_{\mathbf{r}}]\hat{a}_{\mathbf{r}'''}\right) \\
&= \sum_{\mathbf{r}'',\mathbf{r}'''} T_{\mathbf{r}'',\mathbf{r}'''}\left(\hat{a}^\dagger_{\mathbf{r}''}\hat{a}_{\mathbf{r}}\{\hat{a}_{\mathbf{r}'''}, \hat{a}^\dagger_{\mathbf{r}'}\} - \hat{a}^\dagger_{\mathbf{r}''}\hat{a}^\dagger_{\mathbf{r}'}\{\hat{a}_{\mathbf{r}'''}, \hat{a}_{\mathbf{r}}\} \right. \\
&\qquad\qquad \left. + \{\hat{a}^\dagger_{\mathbf{r}''}, \hat{a}^\dagger_{\mathbf{r}'}\}\hat{a}_{\mathbf{r}}\hat{a}_{\mathbf{r}'''} - \{\hat{a}^\dagger_{\mathbf{r}''}, \hat{a}_{\mathbf{r}}\}\hat{a}^\dagger_{\mathbf{r}'}\hat{a}_{\mathbf{r}'''}\right) \\
&= \sum_{\mathbf{r}'',\mathbf{r}'''} T_{\mathbf{r}'',\mathbf{r}'''}\left(\hat{a}^\dagger_{\mathbf{r}''}\hat{a}_{\mathbf{r}}\delta_{\mathbf{r}'''\mathbf{r}'} - \hat{a}^\dagger_{\mathbf{r}'}\hat{a}_{\mathbf{r}'''}\delta_{\mathbf{r}''\mathbf{r}}\right) \\
&= \sum_{\mathbf{r}''}\left(T_{\mathbf{r}'',\mathbf{r}'}\hat{a}^\dagger_{\mathbf{r}''}\hat{a}_{\mathbf{r}} - T_{\mathbf{r},\mathbf{r}''}\hat{a}^\dagger_{\mathbf{r}'}\hat{a}_{\mathbf{r}''}\right).
\end{aligned}
$$

To calculate $[\hat{T}, \hat{a}^\dagger_{\mathbf{r}}\hat{a}_{\mathbf{r}'}]$ we just need to interchange the $\mathbf{r}$ and $\mathbf{r}'$ in the above relation. Thus we have for the fist commutator in (A.11)

$$[\hat{T}, \hat{a}^\dagger_{\mathbf{r}'}\hat{a}_{\mathbf{r}} - \hat{a}^\dagger_{\mathbf{r}}\hat{a}_{\mathbf{r}'}] = \sum_{\mathbf{r}''}\left(T_{\mathbf{r}',\mathbf{r}''}(\hat{a}^\dagger_{\mathbf{r}''}\hat{a}_{\mathbf{r}} + \hat{a}^\dagger_{\mathbf{r}}\hat{a}_{\mathbf{r}''}) - T_{\mathbf{r},\mathbf{r}''}(\hat{a}^\dagger_{\mathbf{r}'}\hat{a}_{\mathbf{r}''} + \hat{a}^\dagger_{\mathbf{r}''}\hat{a}_{\mathbf{r}'})\right). \tag{A.12}$$

Taking the expectation value of this term we find

$$\langle\Psi(t)|[\hat{T}, \hat{a}^\dagger_{\mathbf{r}'}\hat{a}_{\mathbf{r}} - \hat{a}^\dagger_{\mathbf{r}}\hat{a}_{\mathbf{r}'}]|\Psi(t)\rangle = 2\mathrm{Re}\sum_{\mathbf{r}''}\left(T_{\mathbf{r}',\mathbf{r}''}\rho(\mathbf{r}, \mathbf{r}''; t) - T_{\mathbf{r},\mathbf{r}''}\rho(\mathbf{r}', \mathbf{r}''; t)\right) \tag{A.13}$$

where the density matrix is given by Eq. (4.6).

For the next term in Eq. (A.11) we can write

$$
\begin{aligned}
[\hat{V}_{ext}, \hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}}] &= \sum_{\mathbf{r}''} v_{\mathbf{r}''} ([\hat{a}_{\mathbf{r}''}^{\dagger} \hat{a}_{\mathbf{r}''}, \hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}}] \\
&= \sum_{\mathbf{r}''} v_{\mathbf{r}''} \left(\hat{a}_{\mathbf{r}''}^{\dagger} [\hat{a}_{\mathbf{r}''}, \hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}}] + [\hat{a}_{\mathbf{r}''}^{\dagger}, \hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}}] \hat{a}_{\mathbf{r}''} \right) \\
&= \sum_{\mathbf{r}''} v_{\mathbf{r}''} \left(\hat{a}_{\mathbf{r}''}^{\dagger} \hat{a}_{\mathbf{r}} \{\hat{a}_{\mathbf{r}''}, \hat{a}_{\mathbf{r}'}^{\dagger}\} - \hat{a}_{\mathbf{r}''}^{\dagger} \hat{a}_{\mathbf{r}'}^{\dagger} \{\hat{a}_{\mathbf{r}''}, \hat{a}_{\mathbf{r}}\} \right. \\
&\quad \left. + \{\hat{a}_{\mathbf{r}''}^{\dagger}, \hat{a}_{\mathbf{r}'}^{\dagger}\} \hat{a}_{\mathbf{r}} \hat{a}_{\mathbf{r}''} - \{\hat{a}_{\mathbf{r}''}^{\dagger}, \hat{a}_{\mathbf{r}}\} \hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}'''} \right) \\
&= \sum_{\mathbf{r}''} v_{\mathbf{r}''} \left(\hat{a}_{\mathbf{r}''}^{\dagger} \hat{a}_{\mathbf{r}} \delta_{\mathbf{r}''\mathbf{r}'} - \hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}''} \delta_{\mathbf{r}''\mathbf{r}} \right) \\
&= v_{\mathbf{r}'} \hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}} - v_{\mathbf{r}} \hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}},
\end{aligned}
$$

where like before we can interchange $\mathbf{r}$ with $\mathbf{r}'$ to get the second commutator for the external potential. So we have

$$
[\hat{V}_{ext}, \hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}} - \hat{a}_{\mathbf{r}}^{\dagger} \hat{a}_{\mathbf{r}'}] = (v_{\mathbf{r}'} - v_{\mathbf{r}}) \left(\hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}} + \hat{a}_{\mathbf{r}}^{\dagger} \hat{a}_{\mathbf{r}'} \right). \tag{A.14}
$$

By taking the expectation value we find

$$
\langle \Psi(t) | [\hat{V}_{ext}, \hat{a}_{\mathbf{r}'}^{\dagger} \hat{a}_{\mathbf{r}} - \hat{a}_{\mathbf{r}}^{\dagger} \hat{a}_{\mathbf{r}'}] | \Psi(t) \rangle = 2(v_{\mathbf{r}'} - v_{\mathbf{r}}) \mathrm{Re} \rho(\mathbf{r}, \mathbf{r}'; t), \tag{A.15}
$$

where the density matrix is defined by

$$
\rho(\mathbf{r}, \mathbf{r}'; t) = \langle \Psi(t) | \hat{a}_{\mathbf{r}}^{\dagger} \hat{a}_{\mathbf{r}'} | \Psi(t) \rangle. \tag{A.16}
$$

And for the commutator containing the interaction Hamiltonian $\hat{W}$ we have

$$
\begin{aligned}
[\hat{W}, \hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}}] &= \frac{1}{2}\sum_{\mathbf{r}'',\mathbf{r}'''} w_{\mathbf{r}'',\mathbf{r}'''}[\hat{a}_{\mathbf{r}''}^{\dagger}\hat{a}_{\mathbf{r}'''}^{\dagger}\hat{a}_{\mathbf{r}''}\hat{a}_{\mathbf{r}'''}, \hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}}] \\
&= \frac{1}{2}\sum_{\mathbf{r}'',\mathbf{r}'''} w_{\mathbf{r}'',\mathbf{r}'''}\left(\hat{a}_{\mathbf{r}''}^{\dagger}\hat{a}_{\mathbf{r}'''}^{\dagger}[\hat{a}_{\mathbf{r}''}\hat{a}_{\mathbf{r}'''}, \hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}}] + [\hat{a}_{\mathbf{r}''}^{\dagger}\hat{a}_{\mathbf{r}'''}^{\dagger}, \hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}}]\hat{a}_{\mathbf{r}''}\hat{a}_{\mathbf{r}'''}\right) \\
&= \frac{1}{2}\sum_{\mathbf{r}'',\mathbf{r}'''} w_{\mathbf{r}'',\mathbf{r}'''}\left(\hat{a}_{\mathbf{r}''}^{\dagger}\hat{a}_{\mathbf{r}'''}^{\dagger}\left(\hat{a}_{\mathbf{r}''}[\hat{a}_{\mathbf{r}'''}, \hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}}] + [\hat{a}_{\mathbf{r}''}, \hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}}]\hat{a}_{\mathbf{r}'''}\right)\right. \\
&\quad \left. + \left(\hat{a}_{\mathbf{r}''}^{\dagger}[\hat{a}_{\mathbf{r}'''}^{\dagger}, \hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}}] + [\hat{a}_{\mathbf{r}''}^{\dagger}, \hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}}]\hat{a}_{\mathbf{r}'''}^{\dagger}\right)\hat{a}_{\mathbf{r}''}\hat{a}_{\mathbf{r}'''}\right) \\
&= \frac{1}{2}\sum_{\mathbf{r}'',\mathbf{r}'''} w_{\mathbf{r}'',\mathbf{r}'''}\left(\hat{a}_{\mathbf{r}''}^{\dagger}\hat{a}_{\mathbf{r}'''}^{\dagger}\left(\hat{a}_{\mathbf{r}''}\hat{a}_{\mathbf{r}}\{\hat{a}_{\mathbf{r}'''}, \hat{a}_{\mathbf{r}'}^{\dagger}\} + \{\hat{a}_{\mathbf{r}''}, \hat{a}_{\mathbf{r}'}^{\dagger}\}\hat{a}_{\mathbf{r}}\hat{a}_{\mathbf{r}'''}\right)\right. \\
&\quad \left. - (\{\hat{a}_{\mathbf{r}'''}^{\dagger}, \hat{a}_{\mathbf{r}}\}\hat{a}_{\mathbf{r}''}^{\dagger}\hat{a}_{\mathbf{r}'}^{\dagger} + \{\hat{a}_{\mathbf{r}''}^{\dagger}, \hat{a}_{\mathbf{r}}\}\hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}'''}^{\dagger})\hat{a}_{\mathbf{r}''}\hat{a}_{\mathbf{r}'''}\right) \\
&= \frac{1}{2}\sum_{\mathbf{r}'',\mathbf{r}'''} w_{\mathbf{r}'',\mathbf{r}'''}\left(\hat{a}_{\mathbf{r}''}^{\dagger}\hat{a}_{\mathbf{r}'''}^{\dagger}\left(\hat{a}_{\mathbf{r}''}\hat{a}_{\mathbf{r}}\delta_{\mathbf{r}'''\mathbf{r}'} + \hat{a}_{\mathbf{r}}\hat{a}_{\mathbf{r}'''}\delta_{\mathbf{r}''\mathbf{r}'}\right)\right. \\
&\quad \left. - (\hat{a}_{\mathbf{r}''}^{\dagger}\hat{a}_{\mathbf{r}'}^{\dagger}\delta_{\mathbf{r}'''\mathbf{r}} + \hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}'''}^{\dagger}\delta_{\mathbf{r}''\mathbf{r}})\hat{a}_{\mathbf{r}''}\hat{a}_{\mathbf{r}'''}\right) \\
&= \frac{1}{2}\sum_{\mathbf{r}''}\left(w_{\mathbf{r}'',\mathbf{r}'}\left(\hat{a}_{\mathbf{r}''}^{\dagger}\hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}''}\hat{a}_{\mathbf{r}} + \hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}''}^{\dagger}\hat{a}_{\mathbf{r}}\hat{a}_{\mathbf{r}''}\right)\right. \\
&\quad \left. - w_{\mathbf{r}'',\mathbf{r}}\left(\hat{a}_{\mathbf{r}''}^{\dagger}\hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}''}\hat{a}_{\mathbf{r}} + \hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}''}^{\dagger}\hat{a}_{\mathbf{r}}\hat{a}_{\mathbf{r}''}\right)\right).
\end{aligned}
$$

Subtracting the other term in the commutator we get:

$$
[\hat{W}, \hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}} - \hat{a}_{\mathbf{r}}^{\dagger}\hat{a}_{\mathbf{r}'}] = \sum_{\mathbf{r}''}\left(w_{\mathbf{r}'',\mathbf{r}'} - w_{\mathbf{r}'',\mathbf{r}}\right)\left(\hat{a}_{\mathbf{r}''}^{\dagger}\hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}''}\hat{a}_{\mathbf{r}} + \hat{a}_{\mathbf{r}''}^{\dagger}\hat{a}_{\mathbf{r}}^{\dagger}\hat{a}_{\mathbf{r}''}\hat{a}_{\mathbf{r}'}\right), \quad \text{(A.17)}
$$

where we used the fact that $\hat{a}^{\dagger}$ anti-commutes with $\hat{a}^{\dagger}$ and $\hat{a}$ anti-commutes with $\hat{a}$.

Taking the expectation value we find

$$
\langle\Psi(t)|[\hat{W}, \hat{a}_{\mathbf{r}'}^{\dagger}\hat{a}_{\mathbf{r}} - \hat{a}_{\mathbf{r}}^{\dagger}\hat{a}_{\mathbf{r}'}]|\Psi(t)\rangle = 2\mathrm{Re}\sum_{\mathbf{r}''}(w_{\mathbf{r},\mathbf{r}''} - w_{\mathbf{r}',\mathbf{r}''})\rho_2(\mathbf{r}, \mathbf{r}'', \mathbf{r}'; t), \quad \text{(A.18)}
$$

where the two particle density matrix is defined by

$$
\rho_2(\mathbf{r}, \mathbf{r}'', \mathbf{r}'; t) = \langle\Psi(t)|\hat{a}_{\mathbf{r}''}^{\dagger}\hat{a}_{\mathbf{r}}^{\dagger}\hat{a}_{\mathbf{r}''}\hat{a}_{\mathbf{r}'}|\Psi(t)\rangle. \quad \text{(A.19)}
$$

Substituting Eqs. (A.13), (A.15) and (A.18) back into Eq. (A.11) and then into Eq. (A.10) we find the force balance equation for a lattice system (3.8).

Appendix B

Universal NLSE for two interacting particles on a two-site lattice (Hubbard dimer)

Below we present the derivation of the universal NLSE for a system of two interacting spin-1/2 fermions on a two-site lattice. This gives another explicit example illustrating the general structure of nonlinear equations appearing in Sec. III.

Since the dynamics of the triplet state on two-sites is trivial we concentrate on the singlet state. The spatial part of the singlet 2-particle wave function is symmetric with respect to the permutation of coordinates. It has three components, ψ_{11}, ψ_{22}, and $\psi_{12} = \psi_{21}$, describing different distribution of the particles over the sites. Because only the potentials orthogonal to a constant are relevant (see Sec. IV) we assume from the very beginning that $v_1 = -v_2 = v$

The Schrödinger equation Eq. (4.1) for two particles in the singlet state on the two-site lattice takes the form

$$
\begin{aligned}
i\partial_t \psi_{11} &= -\sqrt{2}\, T\psi_{12} + (2v + w_{11})\psi_{11} & \text{(B.1a)} \\
i\partial_t \psi_{12} &= -\sqrt{2}\, T\psi_{11} + w_{12}\psi_{12} - \sqrt{2}\, T\psi_{22}, & \text{(B.1b)} \\
i\partial_t \psi_{22} &= -\sqrt{2}\, T\psi_{12} + (-2v + w_{22})\psi_{22}. & \text{(B.1c)}
\end{aligned}
$$

To construct NLSE for this system we need to substitute the potential from the force balance equation which relates the on-site potential to the density and wave function. As we have a two-site lattice the structure of the force balance equation coincides with that of Eq. (3.24) derived in Sec. IV. We only have to substitute the values of $k_{12} = 2T\text{Re}\rho_{12}$ and $q_{\mathbf{r}}$ calculated for the two particle system into Eq. (3.24) [by means of Eqs. (4.6) and (3.9)]

$$k_{12} = 2T\text{Re}\rho_{12} = 4T\text{Re}[\psi_{11}^*\psi_{12} + \psi_{12}^*\psi_{22}], \tag{B.2}$$

$$
\begin{aligned}
q_1 = \quad & 4T^2(|\psi_{22}|^2 - |\psi_{11}|^2) - 4T\Big(\text{Re}[\psi_{11}^*\psi_{12}](w_{11} - w_{12}) \\
& - \text{Re}[\psi_{22}^*\psi_{12}](w_{22} - w_{12})\Big) = -q_2.
\end{aligned} \tag{B.3}
$$

As we know $\hat{K}$ matrix has the trivial zero eigenvalue corresponding to a constant potential which is already projected out by setting $v = v_1 = -v_2$. If $2T\text{Re}\rho_{12} \neq 0$ we can invert $\hat{K}$ matrix in Eq. (3.24), which determines the potential v in terms of density and the wave function

$$v(n, \psi) = -\frac{\ddot{n}_1 - q_1}{4T\text{Re}\rho_{12}}. \tag{B.4}$$

Finally by substituting v of Eq. (B.4) into Eq. (B.1) we arrive to the universal NLSE for this system

$$
\begin{aligned}
i\partial_t\psi_{11} &= \Big(-\frac{\ddot{n}_1 - q_1}{2T\text{Re}[\psi_{11}^*\psi_{12} + \psi_{12}^*\psi_{22}]} + w_{11}\Big)\psi_{11} \\
&\quad - T\psi_{12},
\end{aligned} \tag{B.5a}
$$

$$i\partial_t\psi_{12} = w_{12}\psi_{12} - T\psi_{11} - T\psi_{22}, \tag{B.5b}$$

$$
\begin{aligned}
i\partial_t\psi_{22} &= \Big(\frac{\ddot{n}_1 - q_1}{2T\text{Re}[\psi_{11}^*\psi_{12} + \psi_{12}^*\psi_{22}]} + w_{22}\Big)\psi_{22} \\
&\quad - T\psi_{12}.
\end{aligned} \tag{B.5c}
$$

We again explicitly see a system of nonlinear ODEs with the nonlinearity of the rational form. As one expects on the general grounds (see Sec III) inclusion of interactions does not introduce any conceptual modification in comparison with the simplest one-particle case considered in Sec. IV. In the subset Ω of the Hilbert space where $\text{Re}\rho_{12} \neq 0$ all the terms in the right hand side of Eqs. (B.5a) and (B.5c) are infinitely differentiable with respect to components $\psi_{\mathbf{rr'}}$ of the wave function. Hence the whole right hand side stays Lipshitz continuous, provided $\psi \in \Omega$. If the density has a continuous second time

derivative $\ddot{n}$, and the initial state $\psi(0) \in \Omega$ is density-consistent in a sense of Eqs. (3.13)–(3.14), then there is a unique solution to NLSE of Eqs. (B.5). Therefore there exists a one-to-one mapping between the density and the potential in the original two-body problem.

Appendix C

Exact Hohenberg-Kohn functional of the two-site Hubbard model by constrained search

In this Appendix we briefly describe how one can construct the exact Hohenberg-Kohn functional (5.12) for two electrons in the two-site Hubbard model by carrying out the constrained search as suggested by Levy [66, 110] and Lieb [111].

The Hilbert space for two fermions on two sites is of dimension six and separates into a singlet and a triplet sector of dimension three each. Since for any value of d, the ground state of $\hat{H}_0 = \hat{T} + \hat{U}$ is a singlet, we may restrict the search in Eq. (5.12) to singlet wavefunctions only. As a basis for the singlet sector we use the eigenstates of Eq. (5.3). Then the most general singlet state may be written as

$$|\Psi\rangle = A_1|g\rangle + A_2|e_1\rangle + A_3|e_2\rangle, \tag{C.1}$$

where we can, without loss of generality, choose the coefficients A_i to be real. In the chosen basis, the expectation value of $\hat{H}_0 = \hat{T} + \hat{U}$ takes the simple form

$$\langle\Psi|\hat{H}_0|\Psi\rangle = \langle\Psi|\hat{T} + \hat{U}|\Psi\rangle = E_g A_1^2 + E_{e_1} A_2^2 + E_{e_2} A_3^2 \tag{C.2}$$

where the eigenvalues E_g, E_{e_1}, and E_{e_2} of the basis functions are given by Eq. (5.5).

The expansion coefficients A_i in Eq. (C.1) are not independent. The normalization condition of the wavefunction $|\Psi\rangle$ leads to

$$A_1^2 + A_2^2 + A_3^2 = 1 . \tag{C.3}$$

In the constrained search we also have to make sure that we are only searching over wavefunctions which yield to a given "density" d. This gives a second condition on the coefficients which reads

$$d = \langle \Psi | \hat{n_1} - \hat{n_2} | \Psi \rangle = 4 \frac{A_1 + \beta_+ A_3}{\sqrt{1 + \beta_+^2}} A_2 \tag{C.4}$$

where β_+ is given by Eq. (5.4). We can use Eqs. (C.3) and (C.4) to eliminate two of the coefficients, say A_2 and A_3, in the constrained search which then becomes a minimization in a single variable, i.e.,

$$F_{\text{HK}}(d) = \min_{A_1} \langle \Psi(A_1, d) | \hat{T} + \hat{U} | \Psi(A_1, d) \rangle . \tag{C.5}$$

In general, this minimization has to be carried out numerically. In Fig. C.1 we show F^{HK} as a function of the dipole moment for various values of U. We note that $F^{\text{HK}}(d)$ is always minimal at $d = 0$. For large values of U the slope of $F^{\text{HK}}(d)$ changes rapidly as one crosses from negative to positive values of d. For vanishing interaction $U = 0$, the minimization can be carried out fully analytically. The resulting functional, the non-interacting kinetic energy, reads

$$T_s(d) = \min_{\Psi \to d} \langle \Psi | \hat{T} | \Psi \rangle = 2T \left(1 - 2\sqrt{1 - \frac{d^2}{8}} \right) . \tag{C.6}$$

Then the Hartree-exchange-correlation energy is given by

$$E_{\text{Hxc}}(d) = F_{\text{HK}}(d) - T_s(d) \tag{C.7}$$

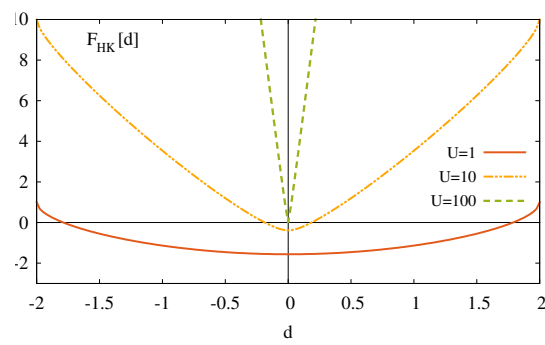and the corresponding Hxc potential can be easily obtained by differentiation.

FIGURE C.1: Exact Hohenberg Kohn density functional $F_{\mathrm{HK}}[d]$ (in units of the hopping parameter T) for different Hubbard strength's U.

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