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Construction of a Kohn-Sham Iteration Scheme in Time-Dependent Lattice Density-Functional Theory

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Abstract

In the present thesis, a rigorous mathematical formulation of time-dependent density-functional theory for lattice systems is derived, serving a formally exact approach towards solving time-dependent many-particle Schrödinger problems. After introducing the necessary mathematical foundations, a one-to-one mapping from external potentials onto electronic densities obtained by solving the Schrödinger equation is introduced. Its properties and the related Banach spaces of scalar potentials and electronic densities are discussed in detail. It allows to precisely formulate the Kohn-Sham Iteration scheme, mapping a given many-particle to an effective single-particle Schrödinger problem, both generating the exact same density. Employing the Banach fixed point theorem, the Kohn-Sham Iteration scheme is proven to be convergent depending on the chosen initial density.

Zusammenfassung

In der hier vorliegenden Arbeit wird ein mathematisches Framework zeitabhängiger Dichte-Funktional-Theorie für quantenmechanische Gitter-Systeme entwickelt. Dies erlaubt einen formal exakten Ansatz für die Lösung der zugehörigen Vielteilchen-Schrödinger-gleichung. Beginnend mit der Diskussion relevanter mathematischer Konzepte, wird eine eins-zu-eins Abbildung zwischen externen Potentialen und zugehörigen, sich aus der Lösung der Schrödingergleichung ergebenden elektronischen Dichten definiert. Selbige, und die zugehörig betrachteten Banachräume externer Potentiale und elektronischer Dichten werden im Hinblick auf die Einführung des sog. Kohn-Sham Iterationsschemas näher untersucht. Das Schema ermöglicht, im Falle von Konvergenz, ein gegebenes interagierendes Vielteilchen-Schrödingerproblem eindeutig auf ein, die selbe Dichte generierendes effektives Einteilchen-Schrödingerproblem zurückzuführen. Unter Verwendung des Banach Fixpunkt Satzes zeigt sich, dass das Kohn-Sham Iterationsschema, abhängig von der gewählten Anfangsdichte, konvergiert.

Viel Freude. Viel Freude. Viel Freude.

Some sticker

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Chapter 1

Introduction

> The most difficult problem ... concerning the use of the language arises in quantum physics. Here we have at first no simple guide for correlating the mathematical symbols with concepts of ordinary language; and the only thing we know from the start is the fact that our common concepts cannot be applied to the structure of the atoms.
>
> Physics and Philosophy: The Revolution in Modern Science,
> Werner Heisenberg

In modern physics, the underlying physical laws of non-relativistic phenomena on microscopic scales are described using the language of Quantum mechanics. Its mathematical formulation relies on the investigation of Cauchy problems on abstract Hilbert spaces [1, 2, 3], i.e. the Schrödinger equation [4]; ubiquitously appearing in various disciplines of science including physics, chemistry, nanotechnology, and biology - only to name a few. From a conceptual point of view, the Schrödinger equation encapsulates the interactions between the constituents of the considered physical system, therefore allowing to determine its physical observables as for instance the bounding energies of molecules, band structures of solids, or even dynamical properties such as the absorption spectrum of an atom. However, considering Schrödinger problems with a large amount of non-decoupling degrees of freedom, i.e. a many-particle Schrödinger problem, calculating exact solutions is not feasible in terms of precision and computational power due to an exponentially increasing dimension of the considered abstract Hilbert space. This is referred to as the *exponential wall problem* [5]. The main object of the present account

is to establish reasonable approximations to many particle Schrödinger problems in order to circumvent the exponential wall problem.

Regarding many-particle systems with Coulombic interaction, time-dependent density-functional theory (TDDFT) is one of the most successful practical methods allowing to describe dynamical electronic properties [6]. It consists of two major conceptual contributions, in principal yielding an exact approach towards solving many-particle Schrödinger problems.

The first conceptual cornerstone of TDDFT was formally justified by Runge and Gross, proving the existence of a one-to-one correspondence between time-analytic external potentials v and the related time-analytic densities n of the many-particle Schrödinger problem [7]. Considering many-particle Schrödinger problems on lattices only, the one-to-one correspondence was established by Farzanehpour and Tokatly for the less restrictive assumption of time-continuous external potentials and in time twice continuously differentiable densities [8]. Employing the one-to-one correspondence, all physical observables can be rephrased as functionals of the related densities, commonly depending on three spacial degrees of freedom, hence allowing to circumvent the exponential wall problem.

The second cornerstone is the existence of the *Kohn-Sham system* [7, 9], defined as the unique non-interacting Schrödinger problem, generating the exact same density of an arbitrary prescribed interacting Schrödinger problem. Both can be identified applying iterative schemes, i.e. the *Kohn-Sham iteration* [10]. Commonly, several reasonable arrpoximations are applied [6], allowing the scheme to converge. In general, however, its mathematical foundations are still not well founded and a formal proof of concept is still missing for Schrödinger problems on lattices.

The following thesis presents a rigorous mathematical formulation of a TDDFT framework for lattice systems, i.e. time-dependent lattice density-functional theory (TDLDFT), and presents a Banach fixed point prove of Kohn-Sham TDLDFT. In the first chapter, the necessary mathematical prerequisites are discussed and a generalization of the Arzela-Ascoli theorem to continuous function spaces of type $\mathcal{C}(I, \mathbb{R}^N)$ is presented (Theorem 2.17). Within the ensuing chapter, a general N particle and M lattice sites many-particle Schrödinger problem is introduced (Equation (3.2)). The many-particle Schrödinger problem is solved formally, allowing to investigate the functional depen-

dence of trajectories on external scalar potentials by means of the potential-trajectory map (Definition 3.2). The results are used to introduce the potential-density map in chapter 4 (Definition 4.1), similarly describing the functional dependence of the density on the external scalar potential.
Chapter 4 is split into three parts. Firstly, the image of the potential-density map is shown to be pre-compact (Corollary 4.4), employing the continuity equation and the theorem of Arzela-Ascoli. Secondly, based on the force balance equation (Equation (4.10)), the Existence theorem of TDLDFT is proved (Theorem 4.11) which states the existence of the Kohn-Sham system restricted to some time of existence (Definition 4.10). The problems of a force balance equation approach towards an Existence theorem are investigated (Section 4.3.2), proving the time of existence to converge to zero for specifc potential configurations. Thirdly, a diffeomorphic mapping property of the potential-density map is established (Theorem 4.14) allowing to introduce the Kohn-Sham Iteration Scheme (Definition 5.3). Employing the diffeomorphic mapping property of the potential-density map, the Kohn-Sham Iteration Scheme is shown to converge using the Banach fixed point theorem (Theorem 5.4), proving the existence of a Kohn-Sham approach towards TDLDFT. Concluding remarks on the time of existence are given.

1.1 Remarks on notation

For convenience, the notion time-dependent density-functional theory is abbreviated by TDDFT and Hartree atomic units will be used.

$$e = \hbar = m_e = \frac{1}{4\pi\epsilon_0} = 1$$

Throughout this thesis, common mathematical notation is used, however, to avoid ambiguity special terminology is explained here.

$\mathbb{N}$	Natural numbers, $\{1, 2, 3, ...\}$
$\mathbb{R}_+$	Real numbers excluding zero
$B_r(x)$	Open Ball with radius r and center point x
$\ell^2(d)$	Hilbert space of square-summable sequences
$\mathcal{C}^k(I, X)$	k-times continuously differentiable function space
$\mathcal{B}(X, Y)$	Banach space of bounded linear operators
$\mathrm{Eig}(\lambda)$	Eigenspace to eigenvalue λ
$\sigma(O)$	Spectrum of an operator O
$f[x]$	Square brackets denote functional dependence
$\|\cdot\|_X$	Norm of Banach space X
$\|\cdot\|_{2,\infty}$	Supremum in time of the $\ell^2(d)$-norm
$\|\cdot\|$	Operator norm of the considered Banach space, usually operator norm of $\ell^2(d)$

Chapter 2

Mathematical Topoi

Within this chapter, the general mathematical concepts, forming the basis of this thesis, are presented. We begin with a formal discussion of non-relativistic quantum mechanical problems by introducing the Banach fixed point theorem and the Schrödinger equation. Hereafter, differentiability on arbitrary Banach space is shown, allowing to state the important inverse function theorem. We conclude with a generalization of the theorem of Arzela-Ascoli to families of continuous functions mapping to arbitrary Euclidean spaces equipped with the Euclidean norm.

2.1 Schrödinger dynamics

This section is dedicated to discuss the dynamics of non-relativistic quantum mechanical problems for arbitrary separable Hilbert spaces $(\mathcal{H}, \langle\cdot,\cdot\rangle_{\mathcal{H}})$ described by the Schrödinger equation. Starting with a review of the Banach fixed point theorem and Banach fixed point theorem iteration, we are able to state the Schrödinger equation as a special case of the abstract Cauchy problem on $\mathcal{H}$ and give conditions for the existence and the uniqueness of solutions. We conclude with an introduction of evolution systems.
In the following, X denotes the Banach space $\big(X, \|\cdot\|_X\big)$.

2.1.1 Banach contraction principle

Within a wide range of physical fields, the concept of fixed points is used to describe several phenomena as equilibria or stability of dynamical systems that are usually formulated by differential equations. Fixed-point schemes allow proving the existence and uniqueness of solutions to these differential equations and are thus of high importance

for physics. One of the most important results regarding the existence of fixed points was found and established by Stefan Banach with the Banach contraction principle, i.e. Banach fixed point theorem [**StefanBanach**].

Definition 2.1 *Let $f : X \to X$ be a mapping.*

1. *An element $x^* \in X$ is called a **fixed point** of f if $f(x^*) = x^*$.*
2. *The map f is called a **contraction** if a non-negative real number $L \in [0, 1)$ exists such that $\|f(x) - f(y)\|_X \leq L \|x - y\|_X \quad \forall x, y \in X$.*

According to Definition 2.1, a contraction f decreases the distance between two points $x, y \in X$. A contraction f maps all points $y \in B_r(x) \subset X$ to a smaller ball $B_{L \cdot r}(f(x))$ with $r \in \mathbb{R}_+$. Iterative application of f to an inital set $B_r(x)$ hence results in a zero sequence of recursively defined radii. This convergent behaviour results in the existence of a fixed point. In the following, we formulate the above statement rigourously and give sufficient conditions for fixed points to exist.

Theorem 2.2 (Banach fixed point theorem) *[Pathak [11], Theorem 5.1]*
*Consider a contraction $f : Y \to Y$ on a closed subset $Y \subseteq X$. Then f admits a unique fixed point. Moreover, the recursively defined sequence $(x_k)_{k \in \mathbf{N}}$ with elements $x_{k+1} \equiv f(x_{k1})$ converges for arbitrary initial values $x_0 \in Y$ to the fixed point. The above iteration scheme is called **Banach iteration scheme**.*

Next, we want to illustrate the Banach fixed point theorem and Banach iteration by investigating the convergence of an algorithm to compute square roots, i.e. the Babylonian method. In the subsequent example, we highlight the importance of both the completeness of the considered set and the contraction property of the mapping.

Example 2.3 *Consider the equation $x^2 - a = 0$ for $a \in \mathbb{R}_+$. We want to find the positive solution to this equation, i.e. the square root of a. By means of the following contraction f, the above equation can be rewritten as a fixed point problem that allows to apply the Banch fixed point theorem.*

$$\begin{array}{rcccl} f & : & [\sqrt{a},\infty) & \longrightarrow & [\sqrt{a},\infty) \\ & & x & \longmapsto & \frac{1}{2}\left(x+\frac{a}{x}\right) \end{array}$$

It can be seen that f is a contraction with Lipschitz constant $L = 1/2$ as for arbitrary $x, y \in [\sqrt{a},\infty)$ the following inequality is satisfied.

$$|f(x) - f(y)| = \frac{1}{2}\left|1 - \frac{a}{xy}\right| |x-y| \leq \frac{1}{2}|x-y|$$

As f is defined on a closed subset $[\sqrt{a}, b]$ of the complete space $\mathbb{R}$ the Banach fixed point theorem can be applied. According to the Banach-iteration scheme the sequence $\left(f(x_{k-1})\right)_{k\in\mathbb{N}}$ with initial value $x_0 = b$ converges to $\sqrt{a}$.

2.1.2 Evolution equations

Let the state space $(\mathcal{H}, \langle\cdot,\cdot\rangle_{\mathcal{H}})$ be an arbitrary separable Hilbert space with $H(t) : D(H(t)) \subseteq \mathcal{H} \to \mathcal{H}$ being a linear self-adjoint map parametically dependend on t for $t \in [0,T]$ with $T \in \mathbb{R}_+$. We define the Hamilton operator H to be the map $t \mapsto H(t)$ and the homogeneous Schrödninger initial value problem as a special case of the abstract Cauchy problem [12]. Since this describes the evolution of a quantum system it is called evolution equation. In the following, we will also refer to this evolution equation by **Schrödinger problem**.

$$\begin{aligned} &\mathrm{i}\tfrac{\mathrm{d}\psi(t)}{\mathrm{d}t} = H(t)\psi(t), \qquad \forall t \in [0,T] \\ &\psi(0) = \psi_0 \in \mathcal{H} \end{aligned} \tag{2.1}$$

We assume that the domain of $H(t)$ is equal to the full state space $D(H(t)) = \mathcal{H}$. This assumption is valid without limitations as we investigate finite dimensional state spaces within this thesis.

The following theorem states the existence of solutions to the Schrödiner inital value problem which are refered to as **trajectories**. It relies on the Banach fixed point theorem and as the theorem is the foundation of this thesis, we present the proof in full detail.

Theorem 2.4 *Consider H to be the Hamilton operator. The Schrödinger problem (2.1) has a unique global, continuously differentiable trajectory ψ, i.e. $\psi \in \mathcal{C}^1([0,T],\mathcal{H})$ if the function $t \mapsto H(t)$ is continuous in the operator norm.*

Proof. This proof follows the idea of the Picard-Lindelöf theorem [12]. Consider X to be the Banach space of continuous functions equipped with the supremum norm, i.e. the function space $\Big(\mathcal{C}([0,T],\mathcal{H}), \|\cdot\|_{\mathcal{H},\infty}\Big)$. Its norm is also denoted by $\|\cdot\|_X$. Based on the mild form of equation (2.1), we define the following map C.

$$\begin{array}{rcccl} C & : & X & \longrightarrow & X \\ & & \psi & \longmapsto & \Big(t \mapsto \psi_0 - i\int_0^t H(\tau)\psi(\tau)\,\mathrm{d}\tau\Big) \end{array}$$

The map C is well-defined since both u and $t \mapsto H(t)$ are continuous functions with a compact domain $[0,T]$. We take the supremum in time of the operator norm and employ continuity of H, thus being bounded by M, i.e $M \equiv \|H\| = \max_{t\in[0,T]} \|H(t)\|$. In order to find a solution to the Schrödinger problem, we calculate

$$\begin{aligned} \|C(\psi) - C(\phi)\|_X &\leq \int_0^T \max_{t\in[0,T]} \Big\| H(\tau)\Big(\psi(\tau) - \phi(\tau)\Big)\Big\|_{\mathcal{H}}\,\mathrm{d}\tau \\ &\leq M\int_0^T \|\psi - \phi\|_X\,\mathrm{d}\tau \\ &\leq M\cdot T\|\psi - \phi\|_X. \end{aligned}$$

Applying the map C twice results in

$$\begin{aligned} \|C^2(\psi) - C^2(\phi)\|_X &\leq \max_{t\in[0,T]} \Big\| \int_0^t \mathrm{d}\tau_2 H(\tau_2)\int_0^{\tau_2} \mathrm{d}\tau_1 H(\tau_1)\Big(\psi(\tau) - \phi(\tau)\Big)\Big\|_{\mathcal{H}} \\ &\leq M^2\cdot\frac{T^2}{2}\|\psi - \phi\|_X. \end{aligned}$$

We estimate the second integral by $M\cdot\tau_2\|\psi - \phi\|_X$ and integrating over the time τ_2 thus results in the fraction $T^2/2$. By principle of induction, this can be generalized to

$$\|C^n(\psi) - C^n(\phi)\|_X \leq M^n\cdot\frac{T^n}{n!}\|\psi - \phi\|_X.$$

Next, we choose $N \in \mathbb{N}$ such that $M^N\cdot\frac{T^N}{N!} < 1$. Therefore, the map C^N is a contraction and by Banach iteration a unique fixed-pont Ψ exists, i.e. $C^N(\Psi) = \Psi$. Applying C yields $C^{N+1}(\Psi) = C(\Psi)$ or equivalently $C^N(C(\Psi)) = C(\Psi)$. By uniqueness of the fixed

point, we can conclude $C(\Psi) = \Psi$, meaning that we also found a fixed point of C satisfying the mild form of (2.1).

$$\Psi(t) = \psi_0 - i \int_0^t H(\tau)\Psi(\tau)\,\mathrm{d}\tau, \quad \forall t \in [0,T] \tag{2.2}$$

By Banach fixed point theorem, the fixed point lies in $\mathcal{C}([0,T],\mathcal{H})$. Therefore, Ψ is a continuous function, meaning that the first derivative of (2.2) exists. Differentiating Ψ results in $i\frac{\mathrm{d}\Psi(t)}{\mathrm{d}t} = H(t)\Psi(t)$ and therefore Ψ is also a solution to the initially stated Schrödinger problem. It is unique in $\mathcal{C}([0,T],\mathcal{H})$ since every solution to the Schrödinger problem is also a solution to the mild form (2.2). □

After solving the Schrödinger equation, we are able to introduce the concept of a time evolution operator $U(t,s)$. It describes the dynamics of a trajectory from a given initial state ψ_0.

Definition 2.5 *Consider a Schrödinger problem (2.1) with the function $t \mapsto H(t)$ being continuous in the operator norm. We define the* ***time evolution operator*** *as the following bounded linear map*

$$\begin{array}{rccc} U(t,0) & : & \mathcal{H} & \longrightarrow & \mathcal{H} \\ & & \psi_0 & \longmapsto & U(t,0)\psi_0 = \psi(t) \end{array} \tag{2.3}$$

where $U(0)\psi_0$ maps to the unique solution of the Schrödinger problem with the initial state $\psi_0 \in \mathcal{H}$.

Following its definition, the time evolution operator can be understood as a unique solution to the following initial value problem.

$$i\frac{\mathrm{d}U(t,0)}{\mathrm{d}t} = H(t)U(t,0), \quad U(0,0) = \mathbb{1}_{\mathcal{H}}, \qquad \forall t \in [0,T]$$

As the Hamilton operator is self-adjoint, $U(t,0)$ is a unitary operator, making it a bijective isometry preserving the norm of trajectories [13]. We generalize the time evolution operator to general times $0 \leq s \leq t \leq T$, describing the time evolution of a state to a given Schrödinger problem from time s to t. We introduce it as $U(t,s) = U(t,0)U^*(s,0)$ and identify the two parameter familiy of time evolution

operators $U(t,s)$ with an evolution system.

Definition 2.6 *A two parameter family of unitary operators $U(t,s)$ on $\mathcal{H}$ for times $0 \leq s \leq t \leq T$ is called an **evolution system** on the Banach space $\mathcal{H}$ if the following conditions are satisfied:*

1. $U(t,t) = \mathbb{1}_X$
2. $U(t,r)U(r,s) = U(t,s) \quad$ *for* $0 \leq s \leq r \leq\leq t \leq T$
3. $U(t,s)^{-1} = U^*(t,s) = U(s,t)$
4. $\partial_t U(t,s) = -iH(t)U(t,s)$

Having introduce the concept of a time evolution operator, we are able to introduce the Heisenberg picture of linear operators acting on the state space $\mathcal{H}$.

Definition 2.7 *The **Heisenberg picture** of the operator $O : \mathcal{H} \to \mathcal{H}$ of a Schrödinger problem (2.1) incorporates the time evolution of the Schrödinger problem and is defined to be*

$$\hat{O}(t) \equiv U(t,0)OU^*(t,0) : \mathcal{H} \to \mathcal{H}, \qquad \forall t \in [0,T].$$

2.2 Differentiability on Banach spaces

Standard calculus and its notion of differentiability only applies to Banach spaces of type $\mathbb{R}^N$. Within this section, we generalize this notion of differentiability to arbitrary Banach spaces. Let $\left(X, \|\cdot\|_X\right)$ and $\left(Y, \|\cdot\|_Y\right)$ be Banach spaces and $f : X \to Y$ define the map of investigation.

2.2.1 Gâteaux and Fréchet differentiability

Definition 2.8 *f is called to be **Gâteaux differentiable** at $x_0 \in X$ if the **Gâteaux derivative** exists at x_0. It is defined as the continuous and linear map*

$$\delta_G f[x_0, \cdot] \quad : \quad X \longrightarrow Y$$

satisfying

$$\lim_{\lambda \to 0} \left\| \frac{f[x_0 + \lambda h] - f[x_0] - \lambda \delta_G f[x_0, h]}{\lambda} \right\|_Y = 0 \tag{2.4}$$

for all $h \in X$ and $\lambda \in \mathbb{R}$. We equivalently denote the Gâteaux derivative by $\delta_G f[x_0]$.

The Gâteaux derivative can be considered as a generalization of the directional derivative in standard calculus. In contrast to the latter, the Gâteaux derivative is defined to be linear in directions $h \in X$. By its definition, the limiting process is required to hold for fixed direction h and thereby enables to compute the Gâteaux derivative by rewriting equation (2.4) as follows.

$$\delta_G f[x, h] = \lim_{\lambda \to 0} \frac{f[x + \lambda h] - f[x]}{\lambda}$$

Generalizing the concept of total differentiability to arbitrary Banach spaces results in introducing the notion of Fréchet differentiability. In comparison to Gâteaux differentiability, Fréchet differentiability requires the limiting process to hold for arbitrary paths $h \to 0$ in X.

Definition 2.9 *f is called to be **Fréchet differentiable** at $x_0 \in X$ if the **Fréchet derivative** exists at x_0. It is defined as the continuous and linear map*

$$D_F f[x_0, \cdot] \quad : \quad X \longrightarrow Y$$

satisfying

$$\lim_{h \to 0} \frac{\|f[x_0 + h] - f[x_0] - D_F f[x_0, h]\|_Y}{\|h\|_X} = 0$$

for all $h \in X$. We equivalently denote the Frèchet derivative by $D_F f[x_0]$. Suppose f is Fréchet differentiable for all $x \in U$ open and the map

$$\begin{aligned} D_F f \quad : \quad U \quad &\longrightarrow \quad \mathcal{B}(X,Y) \\ x \quad &\longmapsto \quad \delta_F f[x,\cdot] \end{aligned}$$

is continuous. Then f is ***continuously differentiable*** *and we write $f \in \mathcal{C}^1(U,Y)$.*

Fréchet differentiability is of great significance in functional analysis as it allows to prove a diffeomorphic mapping property of f. A function f is defined to be a diffeomorphism if f is bijective and both f and its inverse are continuously differentiable. The inverse function theorem states sufficient conditions for f to define a diffeomorphism in an open neighobourhood of one point of its domain.

Theorem 2.10 (Inverse function theorem) *[Pathak [11], Theorem 3.14]*
Consider $U \subseteq X$ to be an open set of Y and $f \in \mathcal{C}^1(U,Y)$. Suppose $x_0 \in U$ such that $D_F f[x_0,\cdot]$ is an isomorphism. Then there exists a neighbourhood $U_x \subseteq U$ of x and $V_x \subseteq Y$ of $y = f(x_0)$ such that $f : U_x \to V_x$ is a diffeomorphism with

$$D_F f^{-1}[y,\cdot] = D_F f[x_0,\cdot]^{-1}.$$

Further properties and rules of calculation for both partial and total differentiablity of standard calculus can be generalized to both Gâteaux and Fréchet differentiability. We are going to use those properties without further discussion. For a more rigorous mathematical treatment see Blanchard-Brüning (2015) [14].

2.2.2 Equivalence of Gâteaux and Fréchet differentiability

In general it is easier to calculate the Gâteaux derivative. In consequence, we want to state an important theorem showing under which conditions both Fréchet and Gâteaux derivative are equivalent.

Theorem 2.11 *[Blanchard-Brüning [14], Lemma 34.3]*
Assume f to be Gâteaux differentiable at all points in an open neighbourhood $U \subseteq X$ of the point $x_0 \in X$ and $x \mapsto \delta_G f[x,\cdot] \in \mathcal{B}(X,Y)$ to be continuous on U. Then f is continuously differentiable, i.e. $f \in \mathcal{C}^1(U,Y)$, with

$$\delta_G f[x_0, h] = D_F f[x_0, h], \quad \forall h \in X.$$

We want to illustrate the difference of both Gâteaux and Fréchet differentiability in more detail, highlighting the significance of Thereom 2.11. We state the following example of a Gâteaux differentiable function that is not Fréchet differentiable.

Example 2.12 *Let f be a function defined by*

$$\begin{array}{rcccl} f & : & \mathbb{R}^2 & \longrightarrow & \mathbb{R} \\ & & x & \longmapsto & f(x) = \begin{cases} \frac{x_1^4 x_2}{\sqrt{x_1^6 + x_2^3}} & , \quad x \neq (0,0) \\ 0 & , \quad x = (0,0) \end{cases} \end{array}$$

The function f is Gâteaux differentiable in $x = 0$ with Gâteaux derivative

$$\delta_G f(0, h) = \lim_{\lambda \to 0} \frac{f(\lambda h)}{\lambda} = \lim_{\lambda \to 0} \lambda \frac{h_1^4 h_2}{\sqrt{\lambda^3 h_1^6 + h_2^3}} = 0$$

for arbitrary $h \in \mathbb{R}^2$. Note that the function f is not continuous in $x = 0$ because the zero sequence $\left(x_n = (n^{-1}, n^{-2})\right)_{n \in \mathbb{N}}$

$$\lim_{n \to \infty} f(x_n) = \frac{1}{2} \neq 0,$$

does not converge to zero. As Fréchet differentiability implies continuity, f cannot be Fréchet differentiable in $x = 0$.

2.3 Theorem of Arzela-Ascoli

Within this section, we prove the theorem of Arzela-Ascoli. It is a generalization of the famous Heine–Borel theorem [15] to continuous function spaces of type $\mathcal{C}(X, \mathbb{R}^N)$ equipped with the supremum norm. In the following, X denotes a metric space $\left(X, d_X\right)$.

The Heine-Borel theorem only applies to finite dimensional vector spaces. It states compactness of any subset of $\mathbb{R}^N$ if and only if it is both closed and bounded. To generalize the Heine-Borel o continuous function spaces of type $\mathcal{C}(X, \mathbb{R}^N)$, we first remind ourselves of its generalization to arbitrary metric spaces X. For this purpose, we introduce the notion of a totally bounded and precompact set X.

Definition 2.13 *A metric space X is **totally bounded** if and only if for any $\epsilon \in \mathbb{R}_+$ there exists a finite cover of X by open balls with radius ϵ.*

Following its definition, any totally bounded set Y is bounded but not vice versa. To illustrate this statement, we state two examples for sets being bounded but not toally bounded.

Example 2.14 *As a first example, note that the closed unit ball in an infinite dimensional Hilbert space cannot be totally bounded. We exemplify this statement by considering the closed unit ball $\overline{B_1(0)}$ of the Hilbert space ℓ^2 which is bounded but not totally bounded. To prove the latter, we assume totally boundedness and proceed by reductio ad absurdum. By totally boundedness, we can construct a finite open cover $C = \left\{B_{1/4}(x) \;\middle|\; x \in Y\right\}$ consisting of balls $\overline{B_1(0)}$ with center points $Y = \{x_1, ..., x_N\}$ and radius $r = 1/4$.*

$$\overline{B_1(0)} \subset \bigcup_{x \in Y} B_{1/4}(x)$$

We consider a Hilbert basis $\{e_i | i \in \mathbb{N}\} \subset \ell^2$. Note that the difference of two different basis vectors is of norm $\sqrt{2}$, i.e $\|e_i - e_j\|_2 = \sqrt{2}$ for any $i \neq j \in \mathbb{N}$ such that each ball $B_{1/4}(x) \in C$ contains at most one basis vector. This contradicts the assumption of totally boundedness, as the Hilbert basis is countably infinite.
We conclude with an example of an infinite but bounded metric space X equipped with the discrete metric d_X. Because of the discrete metric, each point $x \in X$ is isolated, meaning

that there exists a neigbhourhood $B_{r<1}(x)$ of x that only contains x. In consequence, any open cover of X by open balls with radius $r < 1$ necessarily requires the infinite set X to be equalt to the set of center points, contradicting totally boundedness.

It is worth being noted that total boundedness is not equivalent to compactness. X is compact if and only if every open cover of X has a finite cover whereas totally boundedness demands for finite open cover by balls with radius ϵ. For compactness, it is thus also necessary to require completeness, similar to the Heine-Borel theorem.

Lemma 2.15 *[Blanchard-Brüning [14], Lemma 34.3]*
A metric space X is compact if and only if it is complete and totally bounded.

We also remind ourselves of the notion of precompactness of a metric space X. A metric space is defined to be precompact if and only if its closure $\overline{X}$ is compact. Employing Lemma 2.15, any metric space is precompact if and only if totally bounded.
The hereafter stated theorem of Arzela-Ascoli is based on the generalization of the Heine-Borel theorem where the considered metric space is identified with the Banach space $\mathcal{C}(X, \mathbb{R}^N)$ equipped with the supremum norm. Employing continuity allows to reformulate the condition of totally boundedness. The latter is shown to be replaced by the notions of a set $\mathcal{F} \subset \mathcal{C}(X, \mathbb{R}^N)$ being pointwise bounded and equicontinuous.

Definition 2.16 *Let $\mathcal{F} \subset C(X, \mathbb{R}^N)$.*

1. *The family $\mathcal{F}$ is **pointwise bounded** if and only if $\sup_{f\in\mathcal{F}} \|f(x)\| < \infty$ for all $x \in X$. It is said to be **equibounded** if and only if $\sup_{f\in\mathcal{F}} \|f\|_\infty < \infty$.*

2. *The family $\mathcal{F}$ is **equicontinuous** if and only if for every $\epsilon > 0$ there exists a $\delta > 0$ such that $\|f(x) - f(y)\| < \epsilon$ for all $f \in \mathcal{F}$ and all $x, y \in X$ with $y \in B_\delta(x)$.*

The underlying idea for the proof of the Arzela-Ascoli theorem is inspired by Driver (2004) [16]. Note, that we universalize the common formulation the Arzela-Ascoli theorem for continuous function spaces $\mathcal{C}(X, \mathbb{R})$ to $\mathcal{C}(X, \mathbb{R}^N)$. We only prove the left implication of the biconditional and safely skip the right implication. The reader is referred to existing literature on the topic for more details [16].

Theorem 2.17 (Arzela-Ascoli Theorem) *Let X be a compact metric space and $\left(C(X,\mathbb{R}^N),\|\cdot\|_\infty\right)$ the Banach space of continuous functions from X to $\mathbb{R}^N$. Then the family $\mathcal{F}\subset C(X,\mathbb{R}^N)$ is precompact, i.e. totally bounded if and only if $\mathcal{F}$ is pointwise bounded and equicontinuous.*

Proof. ($\Leftarrow$) We first note that any subset of a Banach space is a metric space if equipped with the metric induced by the norm of the Banach space. Hence, we identify $\mathcal{F}$ with the induced metric space and employ Lemma 2.15 to prove for precompactness. In case of precompactness, we demand $\mathcal{F}$'s closure to be compact such that it is sufficient to show that $\mathcal{F}$ is totally bounded. We start proving for equiboundedness as it allows to conclude for $\mathcal{F}$ being totally bounded.
Consider an aribtrary $\epsilon>0$. Employing equicontinuity of $\mathcal{F}$, there exists a $\delta>0$ such that

$$\|f(y)-f(x)\|<\epsilon$$

for all $f\in\mathcal{F}$ and all $x\in X$ with $y\in B_\delta(x)$. Thus any f is constant within to ϵ on $B_\delta(x)$ for all $x\in X$. By compactness of X, we can always construct a finite open cover $C_X=\left\{B_\delta(x)|x\in Z\subset X\right\}$ of X, allowing for the following inequality.

$$\sup_{x\in X}\|f(x)\|\leq\sup_{x\in Z}\|f(x)\|+\epsilon,\qquad\forall f\in\mathcal{F}.$$

We take the supremum over all $f\in\mathcal{F}$ and employ that $\mathcal{F}$ is pointwise bounded. As Z is a finite subset of X, its supremum exists which results in equiboundedness with the constant $M_\epsilon\in\mathbb{R}_+$.

$$\sup_{f\in\mathcal{F}}\|f\|_\infty\leq\sup_{x\in Z}\sup_{f\in\mathcal{F}}\|f(x)\|+\epsilon\leq M_\epsilon$$

Since we consider the Euclidean norm, any component of $f\in\mathcal{F}$ is also bounded by M_ϵ, i.e. $f_i\in[-M_\epsilon,M_\epsilon]$ for all $i\in\{1,...,N\}$, allowing for all functions f to be approximated by a finite amount of values with precision ϵ. To clarify this, we define the finite set of possible values $\mathbb{D}$

$$\mathbb{D}=\left\{\frac{k\epsilon}{N}|k\in\mathbb{Z}\right\}^N\cap[-M_\epsilon,M_\epsilon]^N$$

and introduce the finite valued functions $\phi:Z\to\mathbb{D}$. The set of all functions ϕ is denoted by $\mathbb{D}^Z$. We consider an arbitrary $f\in\mathcal{F}$ and a function $\phi\in\mathbb{D}^Z$. The set of

possible values $\mathbb{D}$ is defined such that we can always choose ϕ satisfying the relation $\|f(x) - \phi(x)\| < \epsilon$ for all $x \in Z$. Employing euqicontinuity, we can deduce that f can indeed be approximated by a function ϕ with precision ϵ.

$$\|f(y) - \phi(x)\| \leq \|f(y) - f(x)\| + \|f(x) - \phi(x)\| < 2\epsilon, \quad \forall x \in Z, y \in B_\delta(x)$$

As f was arbitrary, we can choose at most $|\mathbb{D}|$ different functions ϕ to construct a finite cover of $\mathcal{F}$, i.e. $\mathcal{F} = \cup_{\phi \in \mathbb{D}^Z} \mathcal{F}_\phi$. $\mathcal{F}_\phi$ is defined to be the subset of $\mathcal{F}$ that is approximated by a $\phi \in \mathbb{D}^Z$.

$$\mathcal{F}_\phi = \left\{ f \in \mathcal{F} | \|f(y) - \phi(x)\| < \epsilon, \forall x \in Z, \forall y \in B_\delta(x) \right\}$$

Finally we are able to construct a finite cover of $\mathcal{F}$ by open balls with radius 2ϵ. For a $\phi \in \mathbb{D}^Z$ with $\mathcal{F}_\phi \neq \emptyset$, we can always choose an $f_\phi \in \mathcal{F}_\phi$ such that

$$\|f - f_\phi\|_\infty \leq \sup_{y \in X} \|f(y) - \phi(x)\| + \sup_{y \in X} \|\phi(x) - f_\phi(y)\| < 2\epsilon$$

for all $x \in X$ and $f \in \mathcal{F}_\phi$. From this it follows that any $f \in \mathcal{F}_\phi$ is element of $B_{2\epsilon}(f_\phi)$, meaning $\mathcal{F}_\phi \subset B_{2\epsilon}(f_\phi)$. We conclude with

$$\mathcal{F} = \bigcup_{\phi \in \mathbb{D}^I} \mathcal{F}_\phi \subset \bigcup_{\substack{\phi \in \mathbb{D}^Z, \\ \mathcal{F}_\phi \neq \emptyset}} B_{2\epsilon}(f_\phi),$$

which proves totally boundedness of $\mathcal{F}$ as ϵ was assumed to be arbitrary.

□

Chapter 3

The Many-Particle Problem on a Lattice

In this section, we investigate the quantum dynamics of an interacting electronic many-particle problem on a lattice. Starting with a motivation for quantum mechanics on discrete Hilbert spaces, we introduce and discuss the Schrödinger problem of consideration and formalize the functional dependence of trajectories and observables on scalar potentials.

Quantum mechanics is commonly taught and discussed using continuous position space, i.e. an infinite dimensional separable Hilbert space with specific boundary conditions. This might lead to the problem of unbounded operators such as the kinetic energy operator in free space. In order to circumvent problems of unboundedness we might approximate position space by bounding and discretizing it. Even without problems of unboundedness, discretizing space usually has its origin in numerics.
As an example, we consider a single particle in position space $\mathbb{R}^3$ described by the state space of square-integrable functions $L^2(\mathbb{R}^3)$. We obtain a lattice system with M sites by dividing a compact subset of the position space $\mathbb{R}^3$ into M small bins. We approximate states $\psi \in L^2(\mathbb{R}^3)$ by its mean-values in those respective bins and identify them as elements of the sequence space of square-summable functions $\psi \in \ell^2(M)$. Therefore, any operator is bounded such as the kinetic energy operator which can be approximated by the finite difference Laplacian [17]. Another example is given by a diluted solid with large lattice spacing and tightly bounded electrons. Discarding all inner structure of the atoms and orbitals allows to describe those localized electrons by means of a lattice

Hamilton operator as done by the tight binding Hubbard model [18], also illustrating the significance of lattice models. More generally speaking, lattice models are per se of interest as from a conceptual point of view it is not yet understood whether space and time are continuous or might have a different, maybe a discrete structure, following recent considerations below and around the Planck scale [19].

3.1 The Schrödinger problem

We consider N electrons and a set of M lattice sites described by the index set $I = \{r_1, ..., r_M\}$ with no specific geometry. Both the number of electrons and sites are assumed to be finite but arbitrarily large. The single particle Hilbert space $\mathcal{H}$ of an electron is of finite dimension M representing each site and can be identified with the Hilbert space $\ell^2(M)$ equipped with the standard inner product.

$$\mathcal{H} \cong \ell^2(M), \quad \langle x, y\rangle_{\mathcal{H}} = \sum_{i=1}^{M} x_i^* y_i \quad \forall x, y \in \ell^2(M)$$

We omit spin degrees of freedom as the Hamilton operator of interest is restricted to Coulomb like interactions and acts as an identity operator on the Hilbert space of possible spin configurations.[1]

To accommodate N electrons, we construct the N particle Hilbert space endowed with the induced inner product of the single particle Hilbert space $\mathcal{H}$

$$\mathcal{H}^N = \bigotimes_{i=1}^{M} \mathcal{H} \cong \ell^2(d), \quad \langle x, y\rangle = \prod_{i=1}^{M} \langle x^i, y^i\rangle_{\mathcal{H}} \quad \forall x, y \in \ell^2(d)$$

where the superscript denotes the associated component of the appropriate single particle Hilbert space. By construction, the N particle Hilbert space $\mathcal{H}^N$ is of dimension $d = N \cdot M$. The standard orthonormal basis of $\ell^2(M)$ and $\ell^2(d)$ is defined to be

$$B = \{e_x\}_{x \in I}, \quad B^N = \{e_{\mathbf{x}}\}_{\mathbf{x} \in I^N}. \tag{3.1}$$

[1] The space of possible spin configurations for spin $s = 1/2$ particles is represented by $\mathbb{C}^2$. Thereof the single particle Hilbert space can be identified with the state space $\mathcal{H} = \ell^2(M) \otimes \mathbb{C}^2$ equipped with the inner product $\langle x \otimes s_1, y \otimes s_2\rangle_{\mathcal{H}} = \langle x, y\rangle_{\ell^2(M)} \cdot \langle s_1, s_2\rangle_{\mathbb{C}^2}$.

We use the notation $\mathbf{x} = (x_1, ..., x_N)$. The basis vector e_x, respectively $e_{\mathbf{x}}$, denotes an electronic basis state occupying site x or sites $\mathbf{x}$ and we therefore write

$$\psi = \sum_{\mathbf{x} \in I^N} \psi(\mathbf{x}) e_{\mathbf{x}} = \sum_{x_1,...,x_N \in I} \psi(x_1, ..., x_N) e_{x_1,...,x_N}$$

for an arbitrary electronic N particle state $\psi \in \ell^2(d)$ with $\psi(x_1, ..., x_N) \in \mathbb{C}$.
The Pauli exclusion principle demands a physical realised fermionic state to be anti-symmetric under the interchange of spin and space variables. As we assume the quantum mechanical system to act trivially on spin degrees of freedom, we choose without loss of generality the spacial part of considered wave functions to transform anti-symmetric. In consequence, we restrict the space of physical states to $\ell^2(d)_A$.[2]

$$\ell^2(d) \supset \ell^2(d)_A = \left\{ \psi \in \bigwedge_{i=1}^{N} \ell^2(M) \;\middle|\; \|\psi\|_2 = 1 \right\}$$

Now we are able to state the Schrödinger problem (see Chapter 2.1.2).

$$\begin{aligned} &\mathrm{i}\tfrac{\mathrm{d}\psi(t)}{\mathrm{d}t} = H(t)\psi(t), \quad \forall t \in [0, T] \\ &\psi(0) = \psi_0 \in \ell^2(d)_A \end{aligned} \tag{3.2}$$

We define the many-particle Hamilton operator H as the following linear self-adjoint map depending parametrically on times in the time intervall $[0, T]$ with $T \in \mathbb{R}_+$.

$$\begin{array}{rcccl} H(t) & : & \ell^2(d) & \longrightarrow & \ell^2(d) \\ & & \psi & \longmapsto & \left(T + V(t) + W\right)\psi, \quad \forall t \in [0, T] \end{array}.$$

The operator $V(t)$ denotes the scalar potential and W the many-particle interaction and T the kintetic energy operator, also called hopping operator. In case of a lattice system, we define the kinetic energy operator T as the operator consisting only out of $H(t)$'s off-diagonal elements. It thus describes the transition rate of electrons changing between different sites and we denote this rate from site $x_i \in I$ to site $x_j \in I$ by the matrix element T_{x_i,x_j}. Since T is a self-adjoint operator, the matrix elements have to satisfy the relation $T^*_{x_i,x_j} = T_{x_j,x_i}$. From a physical point of view the matrix elements could be identified with the overlap of Wannier orbitals [20].

[2] $\bigwedge$ denotes the wedge product.

As in the continuous case, the scalar potential $V(t)$ and many-particle interaction W are defined to be real-multiplicative operators. The scalar potential $V(t)$ can be identified with the external potential landscape of the considered system. For a tupel of real scalar potentials $(v(x))_{x\in I}$, with potentials $v(x)$ acting on each site $x \in I$, we define $V(t)$ as follows.[3]

$$V(t) = v(\mathbf{x}, t) = \sum_{i=1}^{N} v(x_i, t), \qquad \forall \mathbf{x} \in I^M, \forall t \in [0, T] \tag{3.3}$$

Within the Hubbard model, this external potential could be identified with a potential generated by the atoms of the considered lattice within the Born-Oppenheimer approximation [20]. The operator W is a two particle operator that is modeling Coulomb like interactions and thus only depends on the spacial distance of two electrons occupying different sites. For two electrons occupying the same site, we set the action of W to zero. Concluding, the Schrödinger problem is thus given by

$$\begin{aligned} i\partial_t \psi(x_1, ..., x_N, t) = &- \sum_{n=1}^{N} \sum_{y_n \in I} T_{x_n, y_n} \psi(..., y_n, ..., t) \\ &+ \sum_{n=1}^{N} v(x_n, t) \psi(x_1, ..., x_N, t) \\ &+ \sum_{i>j}^{N} w(|x_i - x_j|) \psi(x_1, ..., x_N, t) \end{aligned}$$

for an arbitrary initial state $\psi(0) = \psi_0 \in \ell^2(d)_A$.

3.2 Trajectories and potentials

Within this section, we investigate the existence of solutions depending on the set of possible scalar potentials entering the Schrödinger problem (3.2) and discuss the chosen topology of our solution space.

By Theorem 2.4, solutions to the Schrödinger problem exist if both $H(t)$ is bounded and $t \mapsto H(t)$ is continuous in the operator norm. In case of (3.2), the operator $H(t)$ is a bounded linear map, i.e. $H(t) \in \mathcal{B}(\ell^2(d))$ for all $t \in [0, T]$ as any linear map defined

[3] We define $V(t) : \ell^2(d) \to \ell^2(d)$ by $V(t)e_{x_1,...,x_N} = \sum_{i=1}^{N} v(x_i, t)e_{x_1,...,x_N}$ for each $e_{x_1,...,x_N} \in B^N$. We equivalently write $V(t) = \sum_{i=1}^{N} v(x_i, t)$.

on a finite dimensional vector space is bounded. Also, the Hamilton operator H is continuous in time since the expression

$$\|H(t) - H(t')\|^2 \leq \sum_{i=1}^{M} |v(r_i, t) - v(r_i, t')|^2,$$

limits to zero for all $t' \in [0, T]$ and arbitrary sequences $t \to t'$ in $[0, T]$ if we assume continuous scalar potentials $t \mapsto v(r_i, t) \in \mathcal{C}([0, T], \mathbb{R})$. We thus identify the scalar potentials on the lattice with a corresponding vector $v \in \mathcal{C}([0, T], \mathbb{R}^N)$ by setting $v = (v(r_1), ..., v(r_M))^T$.

Definition 3.1 *The Banach space of scalar potentials V is defined to be*

$$V = \left(\mathcal{C}([0, T], \mathbb{R}^M), \|\cdot\|_{2,\infty}\right),$$

such that unique solutions to the Schrödinger problem (3.2) exist for all $v \in V$. The Hamilton operator to a scalar potential $v \in V$ is denoted by

$$H([v], t) = T + V([v], t) + W, \quad \forall t \in [0, T].$$

To highlight the dependence on the multi-particle interaction operator W, we might also write $H[v, W]$. We denote the related Schrödinger problem with an initial state $\psi_0 \in \ell^2(d)_A$ by $\mathcal{S} = \left(\psi_0, v, W\right)$.

We can now regard trajectories as functions of the scalar potential v as we keep T and W fixed. To a given potential, the Hamilton operator $H[v]$ generates an evolution system describing the time evolution starting from an initial state ψ_0. The resulting trajectory will then be denoted as $U[v]\psi_0 : t \mapsto U([v], t, 0)\psi_0$ where the operator $U([v], t, 0)$ denotes the time evolution operator for the assumed potential $v \in V$ (Definition 2.5).

Definition 3.2 *Consider a Schrödinger problem* $\mathcal{S} = \left(\psi_0, v, W\right)$ *with arbitrary scalar potential* $v \in V$*. Its evolution system defines the* ***potential-trajectory map*** *for fixed initial state* ψ_0 *and multi-particle interaction operator* W*.*

$$\begin{array}{rccl} U[\cdot]\psi_0 & : & V & \longrightarrow & X \\ & & v & \longmapsto & \psi[v] = U[v]\psi_0 \end{array} \tag{3.4}$$

X *is defined to be the space of continuous functions* $X = \left(\mathcal{C}([0,T], \ell^2(d)), \|\cdot\|_{2,\infty}\right)$ *equipped with the* $\|\cdot\|_\infty$*-norm, also denoted as* ***trajectory space****.*

Within the rest of this section, we consider a Schrödinger problem $\mathcal{S} = \left(\psi_0, v, W\right)$ and characterize the potential-trajectory map in more detail. Using $\psi[\cdot]$, we can introduce the notion of the so called one-particle density $n([v], x, t)$. For a more detailed discussion we refer to Chapter 4.

$$n([v], x, t) = N \sum_{\mathbf{z} \in I^{M-1}} |\psi([v], x, \mathbf{z}, t)|^2, \qquad \forall x \in I, \forall t \in [0,T]$$

Employing this notion of a one-particle density, we prove for injectivity of the potential-trajectory map $\psi[\cdot]$.

Theorem 3.3 *A trajectory is uniquely defined by a potential* $v \in V$ *if its one-particle density* $n([v], x, t)$ *is assumed to be non-zero for all lattice sites* $x \in I$ *and times* $t \in [0,T]$*.*

Proof. We assume $\psi[v] = \psi[w]$ for $v, w \in V$ and take the difference of both Schrödinger equations, where we employ the equivalence of both trajectories. The potential difference $v - w$ is denoted by $\Delta v = v - w$.

$$0 = V([\Delta v], t)\psi([v], t), \qquad \forall t \in [0,T]$$

We use the explicit basis representation of the trajectory $\psi([v], t)$ and obtain the equivalent condition for its coefficients.

$$\sum_i^N \Delta v(x_i, t)\psi([v], \mathbf{x}, t) = 0, \qquad \forall \mathbf{x} \in I^N, \forall t \in [0,T]$$

We multiply by N and the complex conjugate of the considered state $\psi^*([v], \mathbf{x}, t)$. We keep the first particle position x_1 of the trajectory fixed while summing over its residual degrees of freedom. The sum $N \sum_{\mathbf{z} \in I^{M-1}} |\psi([v], \mathbf{z}, t)|^2$ is identified with the notion of the one-particle density $n([v], x_1, t)$.

$$\sum_{i=1}^{N} \Delta v(x_i, t) n([v], x_1, t) = 0, \qquad \forall \mathbf{x} \in I^N, \forall t \in [0, T]$$

By assumption, $n([v], x_1, t)$ is non-zero for all $x_1 \in I$ and $t \in [0, T]$. Dividing by $n([v], x_1, t)$ and solving for $\Delta v(x_1, t)$ thus results in

$$\Delta v(x_1, t) = -\sum_{i=2}^{N} \Delta v(x_i, t), \qquad \forall \mathbf{x} \in I^N, \forall t \in [0, T]$$

such that the right hand side is constant with respect to x_1. As x_1 is assumed to be arbitrary, this yields that any potential difference $\Delta v(x_i, t)$ has to be a function in time, i.e. $\Delta v(x_i, t) = c(t)$. Substituting $c(t)$ into the above equation yields $c(t) = 0$.

$$\sum_{i=1}^{N} \Delta v(x_i, t) = \sum_{i=1}^{N} c(t) = 0, \qquad \forall \mathbf{x} \in I^N, \forall t \in [0, T]$$

We thus conclude $\Delta v = 0$,i.e. $v = w$ and thus the uniqueness of the trajectory $\psi[v]$. □

We conclude this section with a proof of Fréchet differentiability, starting with uniform continuity, illustrating important proof ideas. We are following the idea of Penz (2016) in Corollary 3.44 [13].

Lemma 3.4 *The potential-trajectory map $\psi[\cdot]$ is uniformly continuous.*

Proof. The potential-trajectory map $\psi[\cdot] = U[\cdot]\psi_0$ is uniformly continuous if

$$\|U[v + h]\psi_0 - U[v]\psi_0\|_{2,\infty} = \sup_{t \in [0,T]} \|U([v + h], t, 0)\psi_0 - U([v], t, 0)\psi_0\|_2$$

goes uniformly for all $v \in V$ to zero for any zero sequence $h \to 0$ in the V-topology. The differences of both trajectories can be rearranged as an integral since $U([v], t, s)$ and $U([v + h], t, s)$ belong to an evolution system (2.6). First, we use property (2.6.1) of evolution systems to rewrite the difference as a product and second, we make use of the fundamental theorem of calculus as the considered time evolution operators are

differentiable in time by property (2.5.4).

$$
\begin{aligned}
&\Big(U([v+h],t,0) - U([v],t,0)\Big)\psi_0 \\
&= U([v+h],t,s)U([v],s,0)\Big|_{s=t}^{s=0}\psi_0 \\
&= -\int_0^t \partial_s U([v+h],t,s)U([v],s,0)\psi_0 \,\mathrm{d}s \\
&= +i\int_0^t U([v+h],t,s)H([v+h],s)U([v],s,0) - U([v+h],t,s)H([v],s)U([v],s,0)\psi_0 \,\mathrm{d}s \\
&= +i\int_0^t U([v+h],t,s)V([h],s)U([v],s,0)\psi_0 \,\mathrm{d}s
\end{aligned}
\tag{3.5}
$$

Next, we estimate the integrand in the $\ell^2(d)$-topology using the unitarity of the time evolution operator $U([v+h],t,s)$.

$$
\|U([v+h],t,s)V([h],s)\psi([v],s)\|_2^2 = \sum_{i=1}^{N} |h(x_i,s)\psi([v],x_i,s)|^2 \leq \|h\|_V^2.
$$

We apply the norm of X Definition 3.2 and obtain

$$
\begin{aligned}
\|U[v+h]\psi_0 - U[v]\psi_0\|_X &\leq \sup_{t\in[0,T]} \int_0^t \Big\|V([h],s)U([v],s,0)\psi_0\Big\|_2 \,\mathrm{d}s \\
&\leq T\|h\|_V
\end{aligned}
$$

Since $h \to 0$ limits to zero in the V-topology, the difference $\|U[v+h]\psi_0 - U[v]\psi_0\|_X$ vanishes uniformly for all $v \in V$ and therefore $U[\cdot]\psi_0$ is uniformly continuous. □

Using uniform continuity, we are able to show continuous differentiability of $\psi[\cdot]$ in form of Theorem 3.5. This is the main result of this chapter.

Theorem 3.5 *The potential-trajectory map $\psi[\cdot]$ is continuously differentiable, i.e. $\psi[\cdot] \in \mathcal{C}^1(V,X)$ with Fréchet derivative $D_F\psi[v,\cdot] : V \to X$ defined by*

$$
D_F\psi[v,w] = \left(t \mapsto -i\int_0^t U([v],t,s)V([w],s)\psi([v],s)\,\mathrm{d}s\right), \quad \forall t \in [0,T].
$$

Proof. The following proof employs Theorem 2.11. It is based on the idea that we can obtain Fréchet differentiability of the potential-trajectory map Definition 3.2, starting with the simpler notion of Gâteaux differentiability. We apply the definition

of the Gâteaux derivative to the potential-trajectory map $\psi[\cdot]$ for arbitrary $h \in X$ and $\lambda \in \mathbb{R}$ (2.8). Following the calculations stated in Lemma 3.4 and performing the same manipulations as in equation (3.5) yields the following equation for the Gâteaux derivative with $\lim_{k\to\infty} \lambda_k = 0$ and $\lambda_k \in \mathbb{R}$.

$$\begin{aligned}\delta_G\psi([v,h],t) &= \lim_{k\to\infty} \frac{\Big(U([v+\lambda h],t,0) - U([v],t,0)\Big)\,\psi_0}{\lambda_k} \\ &= -i \lim_{k\to\infty} \int_0^t U([v+\lambda_k h],t,s)V([h],s)U([v],s,0)\psi_0 \,\mathrm{d}s, \quad \forall t \in [0,T].\end{aligned}$$

Next, we want to apply the dominated convergence theorem in order to interchange the limit $\lim_{k\to\infty} \lambda_k = 0$ and the integral. We thereof define the sequence of functions $(f([v+\lambda_k h]t,s))_{k\in\mathbb{N}}$ with elements

$$f([v+\lambda_k h]t,s) = U([v+\lambda_k h],t,s)V([h],s)\psi([v],s), \quad \forall t,s \in [0,T].$$

All $f([v+\lambda_k h]t,s)$ are uniformly continuous in potentials by Lemma 3.4. Thus, the sequence of functions is uniformly converging to $U([v],t,s)V([h],s)\psi([v],s)$. As all $s \mapsto f([v+\lambda_k h]t,s)$ and the limit value $s \mapsto U([v],t,s)V([h],s)\psi([v],s)$ are integrable, we can apply the dominated convergence theorem.

$$\delta_G\psi([v,h],t) = -i \int_0^t U([v],t,s)V([h],s)U([v],s,0)\psi_0 \,\mathrm{d}s, \quad \forall t \in [0,T] \tag{3.6}$$

In order to identify the map $\delta_G\psi[v,\cdot]$ with the Gâteaux derivative, it has to be linear and continuous. This is satisfied since $V([h])$ is defined to be a multiplicative operator. It also provides continuity as

$$\begin{aligned}\|\delta_G\psi[v,h+w] - \delta_G\psi[v,w]\|_X &= \sup_{t\in[0,T]} \left\| \int_0^t U([v],t,s)V([w],s)\psi([v],s)\,\mathrm{d}s \right\|_2 \\ &\leq T\|w\|_V\end{aligned}$$

limits to zero for any zero sequence $w \to 0$ in the V-topology for arbitrary $v \in V$ Definition 3.1. The first rearrangement used linearity and the second followed the estimation of equation (3.5). Thus $\delta_G\psi[v,\cdot]$ is the Gâteaux derivative of $\psi[\cdot]$.
Now, we show the continuous differentiability of $\psi[\cdot]$ by employing Theorem 2.11. For this purpose, we have to prove continuity of the Gâteaux derivative in scalar potentials $v \in V$ within the $\mathcal{B}(V,X)$-topology, i.e the continuity of the map $v \mapsto \delta_G\psi[v,\cdot] \in \mathcal{B}(V,X)$.

Consider the difference

$$\begin{aligned}
&\delta_G\psi[v+h,w] - \delta_G\psi[v,w] \\
&\quad= \int_0^t \mathrm{d}s U([v+h],t,s)V([w],s)\psi([v+h],s) - U([v],t,s)V([w],s)\psi([v],s) \\
&\quad= \int_0^t \Big(U([v+h],t) - U([v],t)\Big)\, V([w],s)U[v+h]\psi_0 \\
&\qquad\quad + U([v],t)V([w],s)\Big(U[v] - U[v+h]\Big)\,\psi_0\,\mathrm{d}s
\end{aligned}$$

for arbitrary potentials $w \in V$. We estimate the difference of Gâteaux derivatives of scalar potentials $v, v+h \in V$ in the $\mathcal{B}(V,X)$-topology. We make use of the notation $U([x],t,\cdot)$ to denote the map $s \mapsto U([x],t,s)$ for arbitrary potentials $x \in V$.

$$\begin{aligned}
&\|\delta_G\psi[v+h,\cdot] - \delta_G\psi[v,\cdot]\| \\
&\leq \sup_{\|w\|_V=1} \int_0^T \mathrm{d}s \Big\| \Big(U([v+h],t,\cdot) - U([v],t,\cdot)\Big)\, V[w]\psi[v+h]\Big\|_{2,\infty} \\
&\qquad\qquad + \Big\|U([v],t,\cdot)V[w]\Big(\psi[v] - \psi[v+h]\Big)\Big\|_{2,\infty}
\end{aligned}$$

The difference vanishes as $\|h\|_V$ limits to zero since both $\psi[\cdot]$ and $U([\cdot],t)$ which are acting on a state ψ_0 are continuous in the X-topology by Lemma 3.4. More explicitly, consider the first term of the integrand. It can be estimated to be

$$\begin{aligned}
&\Big\| \Big(U([v+h],t,\cdot) - U([v],t,\cdot)\Big)\, V[w]\psi[v+h]\Big\|_{2,\infty} \\
&\leq \|V[w]\| \sup_{\|\psi\|_2=1} \cdot \Big\| \Big(U([v+h],t,\cdot) - U([v],t,\cdot)\Big)\,\psi\Big\|_{2,\infty} \\
&\leq \|V[w]\| \cdot \|h\|_{2,\infty}
\end{aligned}$$

with $\|V[w]\|$ being finite since the potential w is continuous in time. Similarly, the second term vanishes for $\|h\|_V \to 0$.
By Theorem 2.11, $\psi[\cdot]$ is continuously differentiable for all directions $h \in V$ with Fréchet derivative

$$D_F\psi[v,h] = \delta_G\psi[v,h] = \left(t \mapsto -i\int_0^t U([v],t,s)V([w],s)\psi([v],s)\,\mathrm{d}s\right), \quad \forall t \in [0,T].$$

□

Chapter 4

The Potential to Density Map

Within the previous chapter, we formally solved the many-particle Schrödinger problem on a lattice and established the notion of a potential-trajectory map $\psi[\cdot]$ (Definition 3.2). Here, we will use this result to discuss how the potential-trajectory map defines physical quantities, for instance the density and current. We investigate this by introducing the potential-density map $n[\cdot]$ describing the electron density of the considered many-particle problem as a function of its scalar potential. Its image, i.e. the set of physical densities, is proven to be precompact in the space of continuous functions $C(X, \mathbb{R}^N)$ with X denoting the trajectory space (Definition 3.2).
Hereafter, we state the existence of lattice TDDFT by proving the Existence theorem (Theorem 4.11), i.e. the lattice analogue of the extended Runge-Gross theorem [21]. The Existence theorem states that any physical density of a Schrödinger problem can be uniquely generated by any other auxiliary Schrödinger problem with different initial state and multi-particle interaction, restricted to arbitrary small time scales. It also ensures the invertibility of the potential-density map $n[\cdot]$ (Definition 4.1) and thus the existence of its inverse, the density-potential map $v[\cdot]$.

4.1 The density

Throughout this section, we consider a Schrödinger problem $\mathcal{S} = \left(\psi_0, v, W\right)$ (Definition 3.1). A trajectory of an evolution system to $\mathcal{S}$ defines the probability density function $\mathbf{x} \mapsto |\psi([v], \mathbf{x}, t)|^2$ for all times $t \in [0, T]$ and lattice configurations $\mathbf{x} \in I^N$. It describes the probability of finding particle one at position x_1, particle two at position x_2 and so forth. Employing this notion, we can introduce the **one-particle density** $n([v], x)$

for lattice site $x \in I$. It is defined to be the marginal distribution of the probability density function.

$$n([v], x, t) = N \sum_{\mathbf{z} \in I^{N-1}} |\psi([v], x, \mathbf{z}, t)|^2, \qquad \forall x \in I, \forall t \in [0, T]$$

Within the rest of this thesis, we mostly discuss evolution properties of the one-particle density. Note, that its first and second time derivative are well-defined as the one-particle density is twice continuously differentiable in time. For calculating its time derivatives, it is suitable to express the one-particle density as an expectation value of linear operators. Time derivatives can then be calculated by employing the Heisenberg equation of motion, allowing to perform only purely algebraic operations in the form of commutators. To take advantage of this algebraic approach, we introduce the following linear operator $\Pi^i_{x,y}$ with $i \in \{1, ..., N\}$.

$$\begin{array}{rcccc} \Pi^i_{x,y} & : & \ell^2(d) & \longrightarrow & \ell^2(d) \\ & & \psi & \longmapsto & \Pi^i_{x,y}\psi = \mathbb{1}_{\ell^2(M)^{i-1}} \otimes e_x e_y^\dagger \otimes \mathbb{1}_{\ell^2(M)^{N-i}} \psi \end{array}, \qquad \forall x, y \in I \quad (4.1)$$

The operator $\Pi^i_{x,y}$ maps the subspace of the i-th particle Hilbert space $\mathbb{C}e_y$ to $\mathbb{C}e_x$ and acts as an identity operator on the residual $N-1$ single particle Hilbert spaces. For x equals y, we obtain a projection operator denoted by Π^i_x. It projects the i-th particle component of any state ψ on the basis state e_x. For convenience, we might also denote the linear operator acting on the first particle Hilbert space just by $\Pi_{x,y}$ respectively Π_x. Employing the notion of Π_x, we can rewrite the one-particle density as follows.

$$n([v], x, t) = N \sum_{\mathbf{z} \in I^{N-1}} |\psi([v], x, \mathbf{z}, t)|^2 = N \langle \Pi_x \rangle_{\psi([v],t)}. \quad (4.2)$$

To perform the last rearrangement, we identify the marginal distribution with the expectation value of the projection operator Π_x. We sum over all degrees of freedom, while keeping the lattice site of the first particle fixed to x which results in a projection on the basis state e_x of the first single particle Hilbert space.

We generalize this notion of the one-particle density by means of the potential-density map describing the dynamics of the one-particle density for all lattice sites simultaneously.

Definition 4.1 *Consider a Schrödinger problem* $\mathcal{S} = \left(\psi_0, v, W\right)$ *with arbitrary scalar potential* $v \in V$ *(Definition 3.1). Its evolution system defines the* ***potential-density map*** *for fixed initial state* ψ_0 *and multi-particle interaction operator* W*.*

$$\begin{aligned} n[\cdot] \quad : \quad V \quad &\longrightarrow \quad D \\ v \quad &\longmapsto \quad n[v] = \begin{pmatrix} n([v],r_1) \\ \vdots \\ n([v],r_M) \end{pmatrix} = \left(t \mapsto N \begin{pmatrix} \langle \Pi_{r_1} \rangle_{\psi([v],t)} \\ \vdots \\ \langle \Pi_{r_M} \rangle_{\psi([v],t)} \end{pmatrix} \right), \qquad \forall t \in [0,T] \end{aligned}$$

Its image $n[V] \subset D$ *is the set of* ***physical densities*** *and* D *denotes the Banach space of all possible densities*

$$D = \left(\mathcal{C}([0,T], \mathbb{R}^M), \|\cdot\|_{2,\infty}\right).$$

The potential-density map $n[\cdot]$ is defined by an inner product involving generalized trajectories $\psi[\cdot]$ (Definition 3.2). Using the Cauchy-Schwarz inequality, continuous differentiability of $\psi[\cdot]$ implies continuous differentiability of $n[\cdot]$, meaning $n[\cdot] \in \mathcal{C}^1(V, D)$. Applying Theorem 2.11, we can calculate the Fréchet derivatives $D_F n[v, \cdot]$ by means of the Gâteaux derivative (Definition 2.8). We consider an arbitrary site $x \in I$ and time $t \in [0,T]$, use the hermicity of the projector Π_x and insert the Fréchet derivative of a generalized trajectory $D_F\psi[v, \cdot]$ (Theorem 3.5).

$$\begin{aligned} D_F n([v,w],x,t) &= \lim_{\lambda \to 0} N \frac{\langle \Pi_x \rangle_{\psi([v+\lambda w],t)} - \langle \Pi_x \rangle_{\psi([v],t)}}{\lambda} \\ &= 2N \,\mathrm{Re} \langle D_F\psi([v,w],t), \Pi_x \psi([v],t) \rangle \\ &= -2N \int_0^t \mathrm{Im} \langle U([v],t,s) V([w],s) \psi([v],s), \Pi_x \psi([v],t) \rangle \,\mathrm{d}s \end{aligned}$$

Next, we employ the properties of time evolution systems (Definition 2.1.2) and make use of the adjoint of $U([v],t,s)V([w],s)U([v],s,0)$. We introduce the Heisenberg picture of the operators $V([w],s)$ and Π_x to the Schrödinger problem $\mathcal{S}$ (Definition 2.7) and obtain the following form of the Fréchet derivative.

$$\begin{aligned} D_F n([v,w],x,t) &= -2N \int_0^t \mathrm{Im} \langle \hat{V}([w],s,s) \hat{\Pi}_x(t) \rangle_{\psi_0} \,\mathrm{d}s \\ &= iN \int_0^t \langle [\hat{\Pi}_x(t), \hat{V}([w],s,s)] \rangle_{\psi_0} \,\mathrm{d}s, \qquad \forall v, w \in V, \forall t \in [0,T] \end{aligned}$$

Next, we rephrase the integral kernel. We employ the notion of projection operators (4.1) and reformulate the scalar potential operator $\hat{V}([w], s, s)$ (see Definition 3.1, Equation 3.3).

$$\hat{V}([w], s, s) = \sum_{x \in I} \sum_{j=1}^{N} w(x, s) \hat{\Pi}_x^j(s), \qquad \forall s \in [0, T] \tag{4.3}$$

Considering the Fréchet derivative of the full potential-density map $n[\cdot]$, we therefore get the linear Volterra integral equation of the first kind [22]

$$D_F n([v, w], t) = \int_0^t k([v], t, s) w(s) \, \mathrm{d}s, \qquad \forall t \in [0, T] \tag{4.4}$$

with the twice continuously differentiable integral kernel $k[v] : [0, T]^2 \to \mathcal{B}(\mathbb{R}^M)$. It is defined by the matrix elements

$$k([v], t, s)_{x,y} = iN \sum_{j=1}^{N} \langle [\hat{\Pi}_x(t), \hat{\Pi}_y^j(s)] \rangle_{\psi_0}, \qquad \forall t \in [0, T], \forall x, y \in I.$$

where we employed equation (4.3). Physical wise, the integral kernel $k[v]$ can be interpreted as a linear response kernel [23].

4.2 The continuity equation and the set of physical densities

The existence of a non-interacting many-particle quantum system generating the same density to a given interacting many-particle system is at the very heart of TDDFT and a Kohn-Sham construction. In order to specify this connection, we like to characterize the set of physical densities in more detail, starting with the continuity equation.

We consider a Schrödinger problem $\mathcal{S} = \left(\psi_0, v, W\right)$ and investigate its one-particle density for site $x \in I$, i.e. $n([v], x)$ (Definitions 3.1, 4.1). Its first time derivative defines the continuity equation of $n([v], x)$ and is given by the Heisenberg equation of motion for Π_x. We employ that Π_x commutes both with W and $V(t)$ for all times $t \in [0, T]$ and obtain

$$\partial_t n([v], x, t) = iN \langle [T, \Pi_x] \rangle_{\psi([v],t)}. \tag{4.5}$$

We only consider the parts of T with non trivial action on the first particle Hilbert space, as the remaining action commutes with Π_x. This results in the hereafter given equation with $\sum_{y,z\in I} T_{y,z}\Pi_{y,z}$ describing the action of T on the first particle Hilbert space.

$$\partial_t n([v],x,t) = iN \sum_{y,z\in I} T_{y,z}\langle[\Pi_{y,z},\Pi_x]\rangle_{\psi([v],t)} = -2N\sum_{y\in I} \operatorname{Im} T_{y,x}\langle\Pi_{y,x}\rangle_{\psi([v],t)} \tag{4.6}$$

Next, we introduce the important notion of the complex link current, allowing to reformulate the continuity equation.

Definition 4.2 *We define the hermitian* ***complex link current*** *by* $Q[v] : I^2 \to \mathcal{C}^1([0,T],\mathbb{C})$ *with*

$$Q([v],t)_{y,x} = 2NT_{y,x}\langle\Pi_{y,x}\rangle_{\psi([v],t)}, \qquad \forall t\in[0,T].$$

where $\psi[\cdot]$ *is the potential-trajectory map (Definition 3.2). The imaginary part of the complex link current denotes the* ***link current***

$$J[v]_{y,x} = \operatorname{Im} Q[v]_{y,x} \quad \forall x,y\in I.$$

The matrix elements of the complex link current $Q([v],t)_{y,x}$ describe the correlation of particle configurations $\langle\Pi_{x,y}\rangle_{\psi([v],t)}$ for sites $x,y\in I$, weighted by the linking transition rate $T_{y,x}$. It is therefore denoted as complex link current and its definition results in the final form of $n([v],x)$'s **continuity equation**.

$$\partial_t n([v],x,t) = -\sum_{y\in I} J([v],t)_{y,x} \tag{4.7}$$

The continuity equation thus relates the rate of change of the density to the sum of all link currents pointing towards site x. It allows to limit the set of physical densities in more detail. As opposed to a quantum system defined on a continuous state space, continuity of scalar potentials, i.e. $v\in V$, is sufficient for physical densities being twice continuously differentiable, meaning $n[V]\subset\mathcal{C}^2([0,T],\mathbb{R}^N)\subset D$. This is due to the vanishing of the commutator $[V,\Pi_x]$ in the continuity equation (4.5) for a one-particle density for arbitrary lattice site $x\in I$. Thus we can apply the Heisenberg equation

of motion twice and by continuity of $H[v]$ and $\psi[v]$ in time, $t \mapsto \partial_t^2 n([v], x, t)$ is also continuous.

$$\partial_t^2 n([v], x, t) = iN\partial_t \langle [T, \Pi_x] \rangle_{\psi([v],t)} = -N \langle [H([v], t), [T, \Pi_x]] \rangle_{\psi([v],t)}.$$

Moreover, it provides the boundedness of the first time derivative of the density which is an inherent characteristic of discrete quantum mechanics which was discussed by Farzanehpour and Tokatly (2012) [8]. Here, we present a formalized proof.

Theorem 4.3 *The set of physical densities $n[V]$ is uniform Lipschitz continuous in time with universal Lipschitz constant $L = M^{3/2} J_{\max}$ and maximal link current $J_{\max} = 2N \max_{y,x \in I} |T_{y,x}|$.*

Proof. Consider a density $n[v]$ for an arbitrary scalar potential $v \in V$. Its one-particle densities obey the continuity equation with $Q[v]$ denoting the complex link current (Definition 4.2).

$$\partial_t n([v], x, t) = -\sum_{y \in I} J([v], t)_{y,x} = -\sum_{y \in I} \operatorname{Im} Q([v], t)_{y,x}, \qquad \forall x \in I, \forall t \in [0, T].$$

Any link current along a link can be estimated using the Cauchy-Schwarz inequality. We employ the definition of a one-particle density (see Equation (4.2)) and that any physical state is of norm one.

$$\begin{aligned} |J([v], t)_{y,x}| &\leq |Q([v], t)_{y,x}| = 2|T_{y,x}| \cdot |N \langle \Pi_{y,x} \rangle_{\psi([v],t)}| \\ &\leq 2|T_{y,x}| \cdot |N \langle \Pi_y \rangle_{\psi([v],t)}| \cdot |N \langle \Pi_x \rangle_{\psi([v],t)}| \\ &= 2|T_{y,x}| \sqrt{n([v], y, t) n([v], x, t)} \\ &\leq J_{\max} \end{aligned} \qquad \forall x \in I, \forall t \in [0, T]$$

The maximal link current is defined to be $J_{\max} = 2N \max_{y,x \in I} |T_{y,x}|$. In consequence, the continuity equation of a one-particle density can be estimated as follows,

$$|\partial_t n([v], x, t)| \leq M J_{\max}, \qquad \forall x \in I, \forall t \in [0, T],$$

and the generalized mean-value theorem yields Lipschitz continuity for all elements of

the set $n[V]$ with an overall Lipschitz constant $L = M^{3/2} J_{\max}$. The pre factor of $M^{3/2}$ is due to the used Euclidean norm of $\mathbb{R}^M$.

$$\|n([v],t) - n([v],t')\| \leq M^{3/2} J_{\max} |t - t'|, \qquad \forall v \in V, \forall t, t' \in [0,T].$$

□

Following Theorem 4.3, the change in time of a physical density $n \in n[V]$ is always bounded by the universal Lipschitz constant L. Physical-wise, this is based on the finite energy spectrum of lattice systems because of the finite dimensional state space $\ell^2(d)$. A change of a one-particle density is caused by a change of a trajectory component transitioning from one basis state to another one. This transition corresponds to a change in energy that is limited by the finite amount of hopping elements, i.e. the linking transition rates $T_{x,y}$. By continuity of scalar potentials $v \in V$, the trajectory cannot change its energy arbitrarily fast. In consequence, the change of a one-particle density has to be bounded. This inherent characteristic of quantum mechanics on discrete state spaces yields the following very important conclusion of precompactness of $n[V] \subset D$.

Corollary 4.4 *The set of physical densities $n[V]$ is relatively compact in D.*

Proof. To prove precompactness, we can apply the theorem of Arzela-Ascoli because the set of physical densities $n[V]$ is a subset of the continuous function space N equipped with the supremum norm. We therefore prove for equicontinuity and point-wise boundedness. The first follows directly by Theorem 4.3. The latter is a direct consequence of the time evolution being unitary, thus preserving the norm of any initial state. Any one-particle density $n([v],x,t)$ of a physical density can be estimated by $|n([v],x,t)| = \langle \Pi_x \rangle_{\psi_0} \leq 1$ for all $x \in I$ and $t \in [0,T]$. This yields point-wise boundedness of the set $n[V]$.

$$\sup_{t \in [0,T]} \left\{ \|n([v],t)\|_2 \;\middle|\; \forall v \in V \right\} \leq N$$

In consequence, the theorem of Arzela-Ascoli can be applied, proving that $n[V]$ is relatively compact in D. □

To conclude this section, we introduce the concept of v-representable densities. It is an important generalization of the notion of physical densities. The latter are a special case of a set of v-representable densities since they have the initial state ψ_0 and multi-particle interaction operator W fixed.

Definition 4.5 *A density $n \in N$ is called **v-representable** if and only if a Schrödinger problem $\mathcal{S} = \left(\psi_0, v, W\right)$ exists such that the potential-density map reproduces the exact same density $n = n[v]$.*
*A density n is denoted by **non-interacting** or **interacting** v-representable if and only if the multi-particle interaction operator W is set to be zero or non-zero.*

4.3 Existence of lattice TDDFT

We introduce the Existence theorem (Theorem 4.11), i.e. the lattice analogue of the extended Runge-Gross theorem [21]. The Existence theorem states the existence of a unique function that maps between two different Schrödinger problems - both generating the same density which is restricted to arbitrary small time scales. It also implies the invertibility of the potential-density map $n[\cdot]$ (Definition 4.1). We establish the Existence theorem by introducing the force balance equation as one of the fundamental equations of TDDFT, relating physical densities and corresponding scalar potentials. This section refers to Farzanehpour and Tokatly (2012) and aims to point out possible problems emerging for a Kohn-Sham construction [8]. Throughout this section we consider a Schrödinger problem $\mathcal{S} = \left(\psi_0, v, W\right)$ with an arbitrary scalar potential $v \in V$ (Definitions 3.1, 3.1).

We start with an investigation of the relation between densities and potentials. A manifest connection of both notions can be obtained by differentiating the continuity equation for an arbitrary one-particle density $n([v], x)$ (4.7). The second derivative of the one-particle density $n([v], x)$ is denoted by the **force balance equation**.

$$\begin{aligned}\partial_t^2 n([v],x,t) &= -\sum_{y\in I}\partial_t J([v],t)_{y,x} \\ &= 2N\sum_{y\in I}\operatorname{Re} T_{y,x}\langle[H([v],t),\Pi_{y,x}]\rangle_{\psi([v],t)} \\ &= 2N\sum_{y\in I}\operatorname{Re} Q([v],t)_{y,x}[v(y,t)-v(x,t)]+q([v],x,t)\end{aligned} \tag{4.8}$$

First, we simply apply the Heisenberg equation of motion to the continuity equation (4.7) and second, we calculate the commutator, performing similar manipulations as in equation (4.6). Note, that the commutator of $V(t)$ and $\Pi_{y,x}$ can be identified with the real part of the complex link current $Q([v],t)_{y,x}$ times the difference of scalar potentials on the respective sites $v(y,t)-v(x,t)$ (Definition 4.2). This emphasizes a direct relation between the link current and the force balance equation. We denote the residual commutator $[T+W,\Pi_{y,x}]$ by $q([v],x,t)$.

$$q([v],x,t) = -2N\sum_{y\in I}\operatorname{Re} T_{y,x}\left(\langle[T,\Pi_{y,x}]\rangle_{\psi([v],t)}+\langle[W,\Pi_{y,x}]\rangle_{\psi([v],t)}\right) \tag{4.9}$$

Note that equation (4.8) is identified with a force balance equation because from a physical point of view, the first derivative of the current can be interpreted as a force acting on the electrons at lattice site x. In accordance with this, $q([v],x,t)$ describes the internal stress force of the Schrödinger problem [24]. Its first term denotes the lattice analogue of a kinetic force $F([v],x,t)^{\text{kin}}$. It includes the forces related to kinetic effects induced by the hopping rate and contains a symmetric second rank tensor $T([v],t)$ which can be understood as the lattice analogue of the stress tensor of a non-interacting quantum system [25].

$$F([v],x,t)^{\text{kin}} = -N\sum_{y\in I}\operatorname{Re} T_{y,x}\left(\langle[T,\Pi_{y,x}]\rangle_{\psi([v],t)}\right) = -N\sum_{y\in I}T([v],t)_{x,y}$$

The second term of the internal stress force $q([v],x,t)$ is of the same form as $F([v],x,t)^{\text{kin}}$ but with the kinetic energy operator T replaced by the multi-particle interaction operator W. We thus, in analogy to $F([v],x,t)^{\text{kin}}$, identify it with the lattice analogue of the forces related to the interaction effects between the electrons.

$$F([v],x,t)^{\text{int}} = -N\sum_{y\in I}\operatorname{Re}T_{y,x}\left(\langle[W,\Pi_{y,x}]\rangle_{\psi([v],t)}\right)$$

The remaining expression

$$\begin{aligned}F([v],x,t)^{\text{ext}} &= 2\sum_{y\in I}\operatorname{Re}Q([v],t)_{y,x}[v(y,t)-v(x,t)]\\ &= 2\sum_{y\in I}\operatorname{Re}\left\{Q([v],t)_{x,y}-\delta_{x,y}\sum_{z\in I}Q([v],t)_{z,x}\right\}v(y,t)\end{aligned}$$

corresponds to the external forces induced by the scalar potential [24]. We obtain the second equality by rewriting the scalar potential $v(x,t)$. We introduce an additional sum with a Kronecker delta, i.e. $v(x,t)=\sum_{z\in I}\delta_{x,z}v(z,t)$ and interchange the indices z and y. From a physical point of view, the real part of the complex link current can be interpreted as the lattice analogue of the gradient of the scalar potential, in analogy to the notion of a conservative force. Taking all this into account, equation (4.8) can be rewritten as the following balance of forces,

$$\sum_{y\in I}\partial_t J([v],t)_{y,x} = F([v],x,t)^{\text{kin}} + F([v],x,t)^{\text{int}} + F([v],x,t)^{\text{ext}},$$

illustrating its name as force balance equation.

For further investigations, we want to consider the force balance equation for all componentes of a physical density $n[v]$. Therefore we rephrase it by introducing, similar to the continuous case of TDDFT, the lattice Sturm-Liouville operator [8].

Definition 4.6 *The **lattice Sturm-Liouville operator** is defined to be the map $K[v]:[0,T]\to\mathcal{B}(\mathbb{R}^M)$ given by a self-adjoint matrix with the following entries*

$$K([v],t)_{x,y} = 2\operatorname{Re}\left\{Q([v],t)_{x,y}-\delta_{x,y}\sum_{z\in I}Q([v],t)_{x,z}\right\},\qquad \forall t\in[0,T],\forall x,y\in I$$

with the complex link current $Q[v]$ (Definition 4.2). The lattice Sturm-Liouville operator for a specific state $\psi\in\ell^2(d)_A$ is denoted by the self-adjoint linear map $K(\phi)\in\mathcal{B}(\mathbb{R}^M)$ where the trajectory $\psi[v]$ (Definition 3.2) in the complex link current is replaced by the

state ϕ.

We insert the lattice Sturm-Liouville operator in the force balance equation (4.8) and obtain its following representation for the Schrödinger problem $\mathcal{S} = \left(\psi_0, v, W\right)$ with arbitrary scalar potential $v \in V$.

$$K([v],t)v(t) = \partial_t^2 n([v],t) - q([v],t), \qquad \forall t \in [0,T] \tag{4.10}$$

From an intuitive point of view, the force balance equation seems to be suitable to relate two different Schrödiner problems both generating the same density. This is based on the force balance equation linearly depending on physical densities and on the lattice Sturm-Liouville operator directly coupling to the scalar potential v. The first allows to replace the physical density $n[v]$ by an arbitrary density $\tilde{n}[\tilde{v}]$ of a second auxiliary Schrödinger problem $\tilde{\mathcal{S}} = \left(\tilde{\psi_0}, \tilde{v}, \tilde{W}\right)$. The latter enables us to solve for the scalar potential v by inverting the lattice Sturm-Liouville operator. In order for the Schrödinger problem $\mathcal{S}$ to generate the exact same density $\tilde{n}[\tilde{v}]$, we have to uniquely solve for the scalar potential v via the coupled system of equations

$$\begin{aligned} &\text{(i)} \quad K([v],t)v(t) = \partial_t^2 \tilde{n}([\tilde{v}],t) - q([v],t), \\ &\text{(ii)} \quad \mathrm{i}\tfrac{\mathrm{d}\psi(t)}{\mathrm{d}t} = H([v,W],t)\psi(t), \end{aligned} \qquad \forall t \in [0,T]. \tag{4.11}$$

Note, that the density itself enters equation (4.11, i) by means of its second derivative. Thus, generating the exact same physical density $\tilde{n}[\tilde{v}]$ of Schrödinger problem $\tilde{\mathcal{S}}$ requires additional constraints on the given initial state $\psi(0) = \psi_0$. The initial state ψ_0 has to be chosen such that the density of the Schrödinger problem $\mathcal{S}$ and its first derivative match with the corresponding values of the density of the Schrödinger problem $\tilde{\mathcal{S}}$. The values of $n(0)$ and $\partial_t n(0)$ to a given initial stated are denoted by the initial data $I(\psi_0)$.

Definition 4.7 *The tupel $I(\psi_0) = \left((n(0), \partial_t n(0)\right)$ denotes the following initial data of density and its first derivative fitting the initial state ψ_0.*

$$n(x,0) = \langle \Pi_x \rangle_{\psi_0}, \quad \partial_t n(x,0) = -2N \sum_{y \in I} \operatorname{Im} T_{y,x} \langle \Pi_{y,x} \rangle_{\psi_0}, \qquad \forall x \in I$$

We consider the system of coupled equations (4.11) as a starting point for further investigations. We stated the general idea of reducing the problem of relating two Schrödinger problems generating the same density to a problem of unique solutions to a system of coupled equations. There are different approaches to investigate this problem. For instance, there exists a global fixed point approach by Ruggenthaler et al. (2015) [10]. Within the hereafter section, we present a different approach. We reformulate the considered system of coupled equations (4.11) as a non-linear differential equation and employ the theorem of Picard-Lindelöf to prove for uniqueness [12].

4.3.1 The Existence theorem

The very beauty of the force balance equation is reasoned in the lattice Sturm-Liouville operator, allowing for a Picard-Lindelöf theorem based approach of relating two Schrödinger problems $\mathcal{S} = \left(\psi_0, v, W\right)$ and $\tilde{\mathcal{S}} = \left(\tilde{\psi}_0, \tilde{v}, \tilde{W}\right)$. The general idea of this approach is to construct the scalar potential v to the given density $\tilde{n}[\tilde{v}]$, henceforth denoted by $\tilde{n}$, via a point-wise inversion in time of the lattice Sturm-Liouville operator in equation (4.11, i). For this purpose, we restrict the state space $\ell^2(d)_A$ to $\Omega \subseteq \ell^2(d)_A$. It is defined to be the subset of $\ell^2(d)_A$ for which the lattice Sturm-Liouville operator is guaranteed to be invertible (see Definition 4.9). We also replace $\psi[v]$ in $K[v]$ and $q[v]$ (Definition 4.6, Equation (4.9)) by arbitrary states $\psi(t)$ within that subset and obtain

$$v([\tilde{n}], \psi(t), t) = K^{-1}(\psi(t))\Big\{\partial_t^2 \tilde{n}(t) - q(\psi(t))\Big\}, \qquad \forall t \in [0, T]$$

Then we insert the resulting scalar potential $v([\tilde{n}], \psi(t), t)$ into the Schrödinger problem and obtain its related non-linear differential equation. It is denoted by non-linear Schrödinger problem as the Hamilton operator both depends on the states $\psi(t)$. Also note that the non-linear Schrödinger problem depends on the density $\tilde{n}$ itself.

$$\begin{aligned} &\mathrm{i}\tfrac{\mathrm{d}\psi(t)}{\mathrm{d}t} = \Big(T + V([\tilde{n}], \psi(t), t) + W\Big)\,\psi(t), \\ &\psi(0) = \psi_0 \in \Omega \end{aligned} \tag{4.12}$$

Under the right conditions, the Picard-Lindelöf theorem states the existence of unique solutions on the restricted subset Ω and thus the existence of the desired scalar potential v. In the following, we are going to formulate this idea with more mathematical rigour, starting with a discussion about the invertibility of the lattice Sturm-Liouville operator.

We will show that the rank of the lattice Sturm-Liouville operator is at most $M-1$. Note that by Gauge symmetry every diagonal element of $K(\psi)$ with $\psi \in \ell^2(d)_A$ is the negative sum of the related matrix row, meaning

$$K(\psi)_{x,x} = -\sum_{y \neq x, y \in I} K(\psi)_{x,y}$$

such that any space constant vector $c = \sum_{i=1}^{M} e_i$ satisfies the equation

$$K(\psi)c = 0.$$

Thus, any space constant vector c is an element of $K(\psi)$'s kernel, i.e. $\dim \ker K(\psi) \geq 1$. This implies that the lattice Sturm-Liouville operator $K(\psi)$ is not invertible on its domain $\mathbb{R}^M$ for any state. Equivalently, the algebraic multiplicity of the eigenvalue $\lambda = 0$ is at least one. As we want to invert the lattice Sturm-Liouville operator, we only consider vectors $x \in \mathbb{R}^M$ being orthogonal to the space constant vector $c = \sum_{i=1}^{M} e_i$, meaning

$$x \in \mathrm{span}(\{c\})^{\perp} \equiv \left\{ x \in \mathbb{R}^M | \langle x, c \rangle = 0 \right\} \tag{4.13}$$

Therefore, we introduce the orthogonal projection $P : \mathbb{R}^M \to \mathrm{span}(\{c\})^{\perp}$ and its right-inverse $E : \mathrm{span}(\{c\})^{\perp} \to \mathbb{R}^M$ and define the **reduced lattice Sturm-Liouville operator** $K_r(\psi)$ for $\psi \in \ell^2(d)_A$.

$$K_r(\psi) \equiv P \circ K(\psi) \circ E : \mathrm{span}(\{c\})^{\perp} \to \mathrm{span}(\{c\})^{\perp} \tag{4.14}$$

Invertibility of $K(\psi)$ is from this point on equivalent to the invertibility of the reduced lattice Sturm-Liouville operator $K_r(\psi)$. In case of the force balance equation, this means to exclude the purely time-dependent space constant vectors from the space of potentials V. This uniqueness modulo a time-dependent space constant is typical for TDDFT. By gauge symmetry this shift corresponds to adding a complex phase to the wave function that has no influence of the associated density.

Definition 4.8 *We define the Banach space of* ***reduced potentials*** $V_r = \{v \in V \,|\, \forall t \in [0,T] : \langle v(t), c\rangle = 0\}$ *to be the orthogonal complement to the subspace spanned by all space constant potentials equipped with the induced norm of* V*. Analogously, we define* $V_r|_\tau$ *to be the restricted Banach space of reduced potentials* V_r *where we restrict the time domain of its underlying continuous functions to* $[0,\tau] \subseteq [0,T]$*.*

The reduced lattice Sturm-Liouville operator $K_r(\psi)$ is invertible if and only if it is of full rank. The rank explicitly depends on the chosen state $\psi \in \ell^2(d)_A$ and thus on the state space itself. Accordingly, we restrict the state space to the subset of K-invertible states $\Omega \subseteq \ell^2(d)_A$. It is defined to be the set of states for which the reduced lattice Sturm-Liouville operator is of full rank.

Definition 4.9 *Define* $\Omega \subseteq \ell^2(d)_A$ *to be the subset of* ***K-invertible states*** *such that* $\operatorname{rank}(K(\psi)) = M - 1$ *or equivalently* $\dim\ker K(\psi) = 1$ *for* $\psi \in \Omega$*.*

Firstly, it is important to mention that the set of K-invertible states is non-empty. By Farzanehpour and Tokatly (2012) it was shown that any groundstate of an arbitrary Hamilton operator is element of Ω, allowing for the existence of solutions to Schrödinger problems on $\Omega \subseteq \ell^2(d)_A$ [8].
Secondly, the set of K-invertible states Ω is independent of the considered homogeneous Schrödinger inital value problem. It only depends on the kinetic energy operator T. Employing the definition of the reduced lattice Sturm-Liouville operator (4.14), a state ψ is element of Ω if and only if the the reduced lattice Sturm-Liouville operator is of full rank.

$$\psi \in \Omega \leftrightarrow \det K_r(\psi) \neq 0 \tag{4.15}$$

Note that the matrix elements of K_r are linear combinations of complex link currents because K_r is defined via projections acting on K (Definition 4.6). As we replace trajectories $\psi[v]$ by ψ within the complex link current, K_r only depends on the kinetic energy operator T. Thus, keeping T fixed for all considered physical systems and only varying scalar potentials $v \in V_r$ and multi-particle interaction operators W, the set of K-invertible states Ω remains unchanged.
Finally note, by restricting the state space of the non-linear Schrödinger problem to Ω, we change the domain of the Hamilton operator to $\mathrm{dom}H = [0,T] \times \Omega$. This means

that a maximal solution ψ to a given physical density $\tilde{n}$ might only exist within a restricted time interval of $[0, T]$. This is the case if and only if the trajectory reaches the boundary of Ω at some point in time. To ensure the existence of solutions on a closed time interval, we introduce the time of existence.

Definition 4.10 *Consider the non-linear Schrödinger problem (4.12) to a Schrödinger problem $\mathcal{S}$ with physical density $\tilde{n}$. The **time of existence** $\tau_{\tilde{n}}^* \in (0, T]$ defines the time interval $[0, \tau_{\tilde{n}}^*]$ for which a solution of the non-linear Schrödinger problem exists.*

Finally, we can sate the Existence theorem of TDDFT, i.e. the lattice analogue of the well-known extended Runge-Gross theorem [21]. It enables to introduce a rigorous formulation of a possible Kohn-Sham construction (see Chapter 5), which is at the very heart of this thesis. We follow the proof idea of Farzanehpour and Tokatly (2012) [8].

Theorem 4.11 (Existence theorem) *Let $\tilde{\mathcal{S}} = \left(\tilde{\psi}_0, \tilde{v}, \tilde{W}\right)$ be a Schrödinger problem with an initial state $\tilde{\psi}_0 \in \Omega$ and scalar potential $\tilde{v} \in V_r$. Its physical density $\tilde{n}[\tilde{v}]$ is denoted by $\tilde{n}$. Consider the Schrödinger problem $\mathcal{S} = \left(\psi_0, v, W\right)$ with arbitrary scalar potential $v \in V_r$ and initial state $\psi_0 \in \Omega$ with the initial data $I(\psi_0) = I(\tilde{\psi}_0)$. Then, there exists a scalar potential v which is uniquely determined within times $[0, \tau_{\tilde{n}}^*]$ such that $n[v]$ generates the density $\tilde{n}$ in the time interval $[0, \tau_{\tilde{n}}^*]$. The time $\tau_{\tilde{n}}^*$ denotes the time of existence of $\mathcal{S}$ to the density $\tilde{n}$.*

Proof. We want to uniquely solve for the scalar potential v via the system of coupled equations (4.11) involving the physical density $\tilde{n}$ of $\tilde{\mathcal{S}}$.

$$\begin{aligned} &\text{(i)} \quad K([v],t)v(t) = \partial_t^2 \tilde{n}(t) - q([v],t), \\ &\text{(ii)} \quad \mathrm{i}\tfrac{\mathrm{d}\psi(t)}{\mathrm{d}t} = H([v,W],t)\psi(t), \end{aligned} \qquad \forall t \in [0,T] \tag{4.16}$$

For this purpose, we first restrict the state space to the set of K-invertible states Ω (Definition 4.9). This guarantees point-wise invertibility of $K([v],t)$ for all trajectories with initial state $\psi_0 \in \Omega$. Next, we want to solve for the trajectories $\psi[v]$ and invert the lattice Sturm-Liouville operator $K([v],t)$ in the force balance equation (4.16, i). This

yields the scalar potential as a function of the physical density.

$$v([\tilde{n}],\psi,t) = K^{-1}(\psi)\Big\{\partial_t^2\tilde{n}(t) - q(\psi,t)\Big\}, \qquad \forall t\in[0,T] \tag{4.17}$$

We insert the potential $v([\tilde{n}],\psi,t)$ into the Cauchy problem (4.16, ii) and check for the existence of solutions to the resulting non-linear Schrödinger problem on Ω.

$$\begin{aligned} &\mathrm{i}\tfrac{\mathrm{d}\psi(t)}{\mathrm{d}t} = \Big(T + V([\tilde{n}],\psi(t),t) + W\Big)\,\psi(t), \\ &\psi(0) = \psi_0 \in \Omega, \end{aligned} \qquad \forall t\in[0,T] \tag{4.18}$$

By Farzanehpour and Tokatly (2012) it was shown that this non-linear Schrödinger problem has a unique continuous solution ψ up to the time of existence $\tau^*_{\tilde{n}}$ [8]. Inserting the related solution ψ into equation (4.17) yields the demanded continuous scalar potential, i.e. the unique solution of the system of coupled equations (4.16) for a density n. Note that we restricted the considered time interval as ψ is only defined up to its time of existence $\tau^*_{\tilde{n}}$ (Definition 4.10).

$$v|_{\tau^*_{\tilde{n}}} \equiv v([\tilde{n}],t) = K^{-1}([\tilde{n}],t)\Big\{\partial_t^2 n(t) - q([\tilde{n}],t)\Big\}, \qquad \forall t\in[0,\tau^*_{\tilde{n}}]. \tag{4.19}$$

As the scalar $v|_{\tau^*_{\tilde{n}}}$ is a unique solution, the difference of $n[v|_{\tau^*_{\tilde{n}}}]$'s and $\tilde{n}$'s force balance equations is given by

$$\partial_t^2 n([v|_{\tau^*_{\tilde{n}}}],t) - \partial_t^2\tilde{n}(t) = 0, \qquad \forall t\in[0,\tau^*_{\tilde{n}}].$$

Hence, both densities are equivalent as the initial data of both Schrödinger problems (Definition 4.7) is assumed to be equivalent, i.e. $I(\psi_0) = I(\tilde{\psi}_0)$. In consequence, we define the scalar potential v to be any continuous continuation of $v|_{\tau^*_{\tilde{n}}}$ in the set of reduced scalar potentials V_r such that the Schrödinger problem $\mathcal{S} = \Big(\psi_0, v, W\Big)$ generates the physical density $\tilde{n}$ of $\tilde{\mathcal{S}}$ up to a time of existence $\tau^*_{\tilde{n}}$.

□

The Existence theorem is of major importance for an introduction of a Kohn-Sham construction (see Chapter 5). We can always choose the special case of a non-interacting Schrödinger problem $\tilde{\mathcal{S}}$, i.e. with vanishing multi-particle interaction operator $\tilde{W}$. In consequence, an arbitrary interacting Schrödinger problem $\mathcal{S}$ can be mapped to the

non-interacting Schrödinger problem $\tilde{\mathcal{S}}$ generating the exact same density up to some time of existence. This means that we can always reduce the amount of complexity of the initially stated Schrödinger problem as we switch from a system of N coupled to N decoupled ordinary differential equations. For a more detailed discussion see Chapter 6. We want to present another important consequence of the Existence theorem. It also ensures the injectivity of the potential-density map $n[\cdot]$, i.e. lattice analogue of the well-known Runge-Gross theorem [7].

Corollary 4.12 *Consider a Schrödinger problem $\mathcal{S} = \big(\psi_0, v, W\big)$ with fixed initial state $\psi_0 \in \Omega$ and arbitrary scalar potential $v \in V_r$. Then the potential-density map $n[\cdot]$ is injective on*

$$V_r|_{\tau^*} = \Big\{ v|_{\tau^*_{n[v]}} \;\Big|\; v \in V_r \Big\}$$

*which incorporates all scalar potentials $v \in V_r$ with their time domain being restricted by the time of existence $\tau^*_{n[v]}$. The inverse of the potential-density map $n[\cdot]|_{V_r|_{\tau^*}}$ is denoted by the **density-potential map** $v[\cdot]$.*

Proof. The density to the considered Schrödinger problem $\mathcal{S}$ is denoted by $n = n[v]$. We employ the Existence theorem and set the Schrödinger problem $\tilde{\mathcal{S}}$ to be equivalent to $\mathcal{S}$. Then, there exists a time of existence τ^*_n and a scalar potential

$$v|_{\tau^*_n} \equiv v([n],t) = K^{-1}([n],t)\Big\{\partial_t^2 n(t) - q([n],t)\Big\}, \qquad \forall t \in [0,\tau^*_n] \tag{4.20}$$

that uniquely reproduces the density n, i.e. $n([v|_{\tau^*_n}],t) = n(t)$ for times $t \in [0,\tau^*_n]$. Note that the above result is valid for any scalar potential $v \in V_r$ as v was assumed to be arbitrary. By uniqueness of $v|_{\tau^*_{n[v]}}$, we conclude injectivity of $n[\cdot]$ on $V_r|_{\tau^*}$. We denote its inverse by the density-potential map $v[\cdot] : n[V_r|_{\tau^*}] \to V_r|_{\tau^*}$ with its action given by equation (4.20).

□

It is important to note that both the Existence theorem (Theorem 4.11) and Corollary 4.12 employ different times of existence. Within the Existence theorem, we relate a Schrödinger problem $\mathcal{S}$ to an auxiliary Schrödinger problem $\tilde{\mathcal{S}}$. In consequence, the stated time of existence $\tau^*_{\tilde{n}}$ of $\mathcal{S}$ depends on the physical density $\tilde{n}$ of $\tilde{\mathcal{S}}$. In case of Corollary 4.12, we take the Schrödinger problem $\mathcal{S}$ as its own reference system,

therefore τ_n^* can be identified with the existence time to its own physical densities. This distinction is of major importance because the first allows to relate different Schrödinger problems and the latter for injectivity. Employing both notions yields the following corollary.

Corollary 4.13 *Consider two Schrödinger problems* $\mathcal{S} = \left(\psi_0, v, W\right)$ *and* $\tilde{\mathcal{S}} = \left(\tilde{\psi}_0, \tilde{v}, \tilde{W}\right)$ *with arbitrary scalar potentials* $v, \tilde{v} \in V_r$ *and intitial states* $\psi_0, \tilde{\psi}_0 \in \Omega$ *with* $I(\psi_0) = I(\tilde{\psi}_0)$. *The related times of existence are denoted by* τ^* *and* $\tilde{\tau}^*$. *We define the* ***common time of existence*** *to a given scalar potential* v,

$$\tau_v^* \equiv \min\{\tau_{n[v]}^*, \tau_{\tilde{n}[v]}^*, \tilde{\tau}_{\tilde{n}[v]}^*, \tilde{\tau}_{n[v]}^*\},$$

such that both potential-density maps $n[\cdot]$ *and* $\tilde{n}[\cdot]$ *define a bijection on the set of scalar potentials*

$$\mathcal{V} = \left\{v|_{\tau_v^*} \,|\, v \in V_r\right\}$$

with identical image $\mathcal{D} = n[\mathcal{V}] = \tilde{n}[\mathcal{V}]$, *meaning that* $\mathcal{D}$ *is* v*-representable for both Schrödinger problems* $\mathcal{S}$ *and* $\tilde{\mathcal{S}}$ *(Definition 4.5).*

Proof. First, note that both potential-density maps $n[\cdot]$ and $\tilde{n}[\cdot]$ are well-defined on the domain of scalar potentials $\mathcal{V}$ as it consists of continuous functions defined on compact time intervals $[0, \tau_v^*] \subseteq [0, T]$. Next, note that the common time of existence τ_v^* is bounded by the times of existence of both Schrödinger problems to its own physical densities, i.e.

$$\tau_{n[v]}^*, \tilde{\tau}_{\tilde{n}[v]}^* \geq \tau_v^*, \qquad \forall v \in V_r.$$

We can therefore employ Corollary 4.12 and prove $n[\cdot]$ and $\tilde{n}[\cdot]$ to be injective on $\mathcal{V}$. To prove for $n[\mathcal{V}]$ to be identical to $\tilde{n}[\mathcal{V}]$, note that τ_v^* is also bounded by the maximal time of existence of Schrödinger problem $\tilde{\mathcal{S}}$ to any density of $\mathcal{S}$ and vice versa, i.e.

$$\tau_{\tilde{n}[v]}^*, \tilde{\tau}_{n[v]}^* \geq \tau_v^*, \qquad \forall v \in V_r.$$

By the Existence theorem, both Schrödinger problems create the exact same densities which yields $n[\mathcal{V}] = \tilde{n}[\mathcal{V}]$.

□

4.3.2 The set of K-invertible states and the time of existence

Within this section, we investigate the relation of the time of existence and the set of K-invertible states in more detail (Definitions 4.10, 4.9). We explicitly state necessary conditions for the invertibility of the lattice Sturm-Liouville operator (Definition 4.6). We proceed with an investigation of a common non-zero time of existence for all scalar potentials (Definition 4.10) to a given initial state and multi-particle interaction operator and discuss why neither the Existence theorem (Theorem 4.11) nor injectivity of the potential-density map (Corollary 4.12) can be established for all densities being defined on a uniform non-zero time domain.

The set of K-invertible states Ω is defined to be the set of states for which the lattice Sturm-Liouville operator $K(\psi)$ is of constant rank $M-1$ or, respectively, the reduced lattice Sturm-Liouville operator $K_r(\psi)$ of full rank (see Equation (4.14)).

$$\psi \in \Omega \leftrightarrow \det K_r(\psi) \neq 0$$

By restricting the discussion to K-invertible states only, solutions to the related non-linear Schrödinger might not be global, meaning that the time of existence is smaller than T. The latter is caused by a lower rank of $K(\psi)$ as a consequence of linear dependence or vanishing of columns or rows. To investigate this issue in more detail, we relate the lattice Sturm-Liouville operator and the continuity equation (4.7). The first is defined to be the real part of a linear combination of the matrix elements of the complex link current $Q(\psi)$,

$$K(\psi)_{x,y} = 2\,\mathrm{Re}\left\{ Q(\psi)_{x,y} - \delta_{x,y} \sum_{z \in I} Q(\psi)_{x,z} \right\}, \qquad \forall x, y \in I,$$

whereas the link current $J(\psi)$ of the continuity equation equals the imaginary part of the complex link current (Definition 4.2). Note that we replace the trajectory $\psi[v]$ in $J[v]$ by an arbitrary state $\psi \in \Omega$ and thus write $J(\psi)$ instead of $J[v]$. We identify the off-diagonal elements of both $K(\psi)$ and $J(\psi)$ by means of

$$K(\psi)_{x,y} = \mathrm{Re}\, Q(\psi)_{x,y}, \quad J(\psi)_{x,y} = \mathrm{Im}\, Q(\psi)_{x,y}, \qquad x \neq y \in I$$

We rewrite the matrix elements of the complex link current via its real and imaginary part and take its squared norm, resulting in the following constraint on the matrix elements of the lattice Sturm-Liouville operator.

$$|Q(\psi)_{x,y}|^2 = |J(\psi)_{x,y}|^2 + |K(\psi)_{x,y}|^2, \qquad x \neq y \in I \tag{4.21}$$

By the above decomposition of the complex link current, we can deduce that $K(\psi)_{x,y}$ is zero if and only if $|Q(\psi)_{x,y}| = |J(\psi)_{x,y}|$. With this in mind, consider a fixed site $x_0 \in I$ and assume $|Q(\psi)_{x_0,y}| = |J(\psi)_{x_0,y}|$ for all sites $y \neq x_0 \in I$. As all $\operatorname{Re} Q(\psi)_{x_0,y}$ are vanishing, the related diagonal matrix element $K(\psi)_{x_0,x_0}$ is also calculated to be zero.

$$K(\psi)_{x_0,x_0} = -2 \sum_{z \neq x_0} \operatorname{Re} Q(\psi)_{x_0,z} = 0$$

In consequence, a complete matrix row of $K(\psi)$ is vanishing, meaning that the rank of the lattice Sturm-Liouville operator is at most of $M-2$. We thus conclude that a state ψ is not an element of Ω if all off-diagonal link currents $J(\psi)_{x_0,y}$ are of the same norm as the complex link current $Q(\psi)_{x_0,y}$ for all $y \neq x_0$.

Another necessary condition for ψ to be element of Ω is the non-vanishing of its one-particle densities. As we consider a specific state ψ, we replaced trajectories $\psi[v]$ by ψ and denote the one particle density by $n(\psi, x)$ (see Equation (4.2)). If we assume $n(\psi, x)$ to be zero,

$$n(\psi, x) = \sum_{\mathbf{z} \in I^{M-1}} |\psi(x, \mathbf{z})|^2 = \langle \Pi_x \rangle_\psi = 0, \tag{4.22}$$

implying $\psi(x, \mathbf{z})$ to be zero for $\mathbf{z} \in I^{M-1}$. In consequence, the complex link current $Q(\psi)_{x,y}$ vanishes for all $y \in I$,

$$Q(\psi)_{x,y} = T_{x,y} \langle \Pi_{x,y} \rangle_\psi = T_{x,y} \sum_{\mathbf{z}, \tilde{\mathbf{z}} \in I^{M-1}} \psi^*(x, \mathbf{z}) \psi(y, \tilde{\mathbf{z}}) = 0,$$

implying that the rank of $K(\psi)$ has to be at most $M-2$.

We want to state an example for the above discussion. It illustrates that a vanishing density component $n(\psi, x) = 0$ is not a sufficient condition for a state ψ to be element of Ω. For this purpose, we consider the following example of a Schrödinger

problem $\mathcal{S} = \left(\psi_0, v, W\right)$ with an initial state $\psi_0 \in \Omega$ and scalar potential $v \in V_r$ (Definition 4.8). We choose both ψ_0 and v such that the time evolution of the given initial state $\psi_0 \in \Omega$ reaches the boundary of Ω by reason of its current being of the same norm as the complex link current. The density is shown to be non-zero within the whole time interval.

Suppose we are only considering a one particle problem and a two dimensional state space, meaning $N = 1$ and $M = 2$ with state space $\ell^2(2) \cong \mathbb{C}^2$. The most general Hamilton operator is of the form

$$H([v],t) = T + V([v],t) = \begin{pmatrix} v(1,t) & T_{1,2} \\ T_{1,2}^* & v(2,t) \end{pmatrix}, \qquad \forall t \in [0,T] \tag{4.23}$$

with hopping rate $T_{1,2} \in \mathbb{C}$ and scalar potential $v = v(1)e_1 + v(2)e_2 \in V = C([0,T],\mathbb{R}^2)$ (Definition 3.1). We consider the corresponding lattice Sturm-Liouville operator, reading

$$K([v],t) = \begin{pmatrix} -K([v],t)_{1,2} & K([v],t)_{1,2} \\ K([v],t)_{1,2} & -K([v],t)_{1,2} \end{pmatrix}, \qquad \forall t \in [0,T] \tag{4.24}$$

with $K([v],t)_{1,2} = 2\,\mathrm{Re}\,T_{1,2}\langle\Pi_{1,2}\rangle_{\psi([v],t)}$. In agreement with the discussion of Section 4.3.1, the kernel of $K([v],t)$ is at least one dimensional as two rows of $K([v],t)$ are linearly dependent such that the space constant vector $c = e_1 + e_2$ is an eigenvector with eigenvalue zero. We thus identify the set of reduced potentials V_r by

$$V_r = \left\{ \begin{pmatrix} -w \\ w \end{pmatrix} \;\middle|\; w \in C([0,T],\mathbb{R}) \right\}$$

and set $v(1,t) = -v(2,t) = t$ for all $t \in [0,T]$. To choose the initial state ψ_0, we first characterize $\Omega \in \mathbb{C}^2$ explicitly. The set of K-invertible states Ω is characterized by the non-vanishing determinant of the reduced lattice Sturm-Liouville operator $K_r(\psi)$ (see Equation (4.14)). The operator is identified to be $K_r(\psi) = -k(\psi)_{1,2} = 2\,\mathrm{Re}\,T_{1,2}\langle\Pi_{1,2}\rangle_\psi$, implying that

$$\psi \in \Omega \leftrightarrow \mathrm{Re}\,T_{1,2}\langle\Pi_{1,2}\rangle_\psi \neq 0$$

To ensure $\psi_0 \in \Omega$, we choose the initial state $\psi_0 = (e_1 - e_2)/\sqrt{2}$, i.e. the ground-state of $H([v],0)$. The resulting time evolution with fixed hopping rate $T_{1,2} = 0.5$ and maximal time $T = 5$ is shown in Figure 4.1.

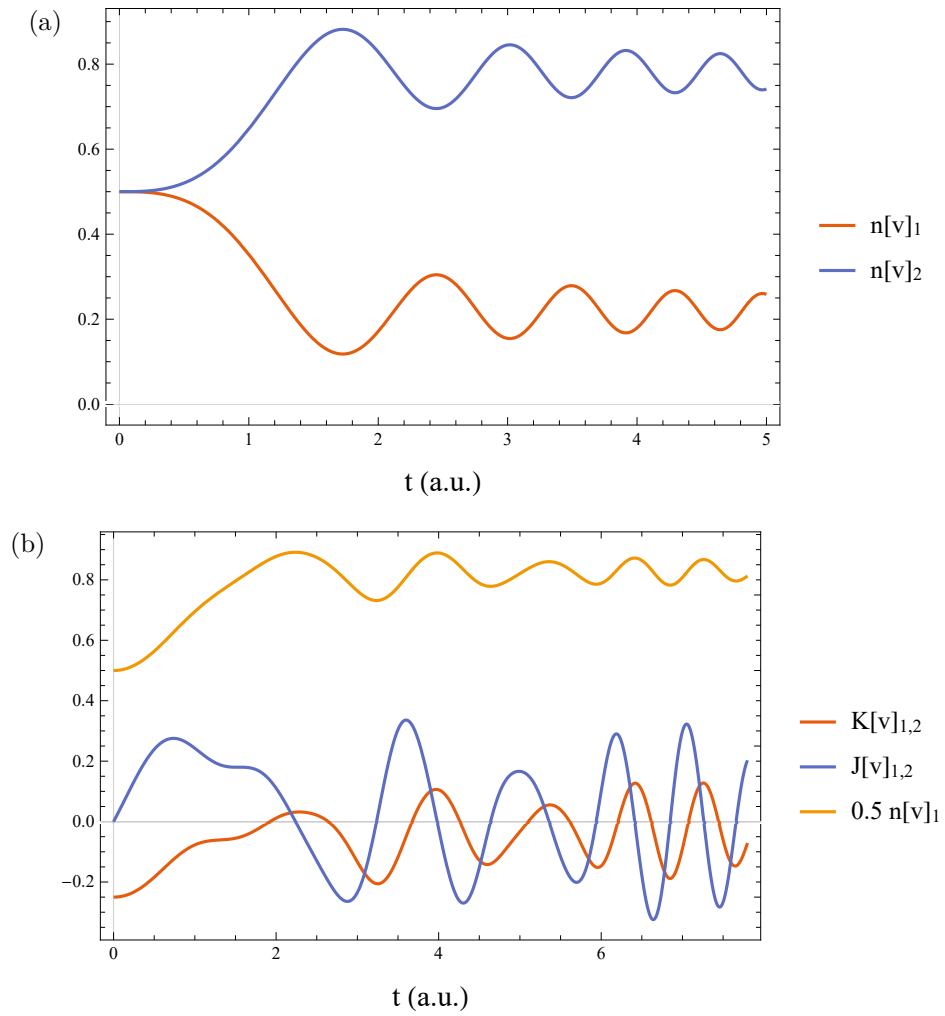

Fig. 4.1: We consider a $N = 1$ particle, $M = 2$ site Schrödinger problem with the initial state $\psi_0 = (e_1 - e_2)/\sqrt{2}$ and Hamilton operator $H[v]$ (4.23). The hopping rate is set to be $T = 0.5$ and scalar potential $v(1,t) = -v(2,t) = t$ for all $t \in [0,5]$.
(a) Time evolution of the density $n([v],1)$ and $n([v],2)$ on site one and two.
(b) Time evolution of the real and imaginary part of the complex link current $Q[v]_{1,2}$, i.e. $K[v]_{1,2}$ and $J[v]_{1,2}$.

Plot (a) presents the time evolution of the electron density for lattice site one and two, i.e. $n([v],1)$ and $n([v],2)$ and plot (b) the related matrix element of the lattice Sturm-Liouville operator $K[v]_{1,2}$ and the current $J[v]_{1,2}$. Considering plot (a), both densities are non-zero within the considered time interval. The electron density $n([v],1)$ decreases within the first moments of time, as the potential acting on site one increases linearly starting with small values. With increasing time, the hopping rate $T_{1,2}$ gets small compared to the spectrum of $H([v],t)$, i.e. $|T_{1,2}/\epsilon| \ll 1$ for all energies $\epsilon \in \sigma(H)$. The kinetic energy operator therefore acts as a small perturbation to the scalar potential $V([v],t)$. In consequence, the trajectory oscillates with a decreasing amplitude around a linear combination of the eigenvectors of $V([v],t)$. Note that the magnitude of

occupation of each site severely depends on the hopping rate $T_{1,2}$. As long as the hopping is of the same magnitude as the energy spectrum, the effective amount of electrons is increasing on site two due to its lower potential compared to site one.
By the continuity equation, the current $J[v]_{1,2}$ reaches its local maximum for any inflection point of $n([v],1)$ and $n([v],2)$. In agreement with equation (4.21) the matrix element of the lattice Sturm-Liouville operator turns zero. In consequence, the trajectory $\psi[v]$ reaches the boundary of Ω such that the time of existence τ_n^* has to be necessarily smaller than $T = 5$ and is $\tau_n^* = 1.308$ for the given potential v.

4.3.3 The uniform time of existence

Consider a Schrödinger problem $\mathcal{S}$ with fixed initial state $\psi_0 \in \Omega$ (Definition 4.9) and multi-particle interaction operator W. We keep its scalar potential $v \in V_r$ (Definition 4.8) arbitrary and investigate the possibility of a common non-zero time scale of existence, i.e. **the uniform time of existence**

$$\tau_{\min} \equiv \inf_{v \in V_r} \left\{ \tau^*_{n[v]} \right\} > 0, \tag{4.25}$$

employing both necessary conditions derived within the last section. Following the discussion of the one-particle density (see Equation (4.2)), a state ψ is element of the K-invertible states Ω if all one-particle density components are non-zero (see Equation (4.2)). As the initial state $\psi_0 \in \Omega$ fixes the initial density (Definition 4.7), all one-particle density components are ensured to be non-zero for $t = 0$. By uniform Lipschitz continuity of the set of physical densities (Theorem 4.3), the absolute value of any one-particle density can change in time at most with the Lipschitz constant $MJ_{\max}$

$$|\partial_t n([v],x,t)| \leq MJ_{\max}, \qquad \forall x \in I, \forall t \in [0,T]$$

such that the $n([v],x)$ cannot vanish arbitrarily fast. In consequence, a vanishing of a one-particle density does not imply $\tau_{\min} = 0$.
Note that a non-vanishing one-particle density is only a necessary condition for ψ to be element of Ω. The rank of the lattice Sturm-Liouville operator also decreases if all link currents pointing towards a specific site are of the same norm as the complex link current (see Equation (4.21)). This is illustrated in the example presented in Figure 4.1.

Following its discussion, we find that the oscillation frequency of $K[v]_{1,2}$ increases in time as the potential difference on site one and two is increasing. With higher oscillation frequency, the distance of consecutive roots of $K[v]_{1,2}$ decreases which suggests that the time of existence $\tau^*_{n[v]}$ to a given Schrödinger problem also decreases with growing potential differences. In consequence, the related common time scale of existence τ might turn zero. To present this argument with more mathematical rigour, we consider the Schrödinger problem $\mathcal{S} = \left(\psi_0, \lambda v, W\right)$ with scalar potential $v \in V_r$ scaled by factor $\lambda \in \mathbb{R}_+$. We assume all components of the scalar potential to be non-zero and to pairwise differ from each other. Its Hamilton operator is given by

$$H([\lambda v], t) = \lambda V([v], t) + T + W, \qquad \forall t \in [0, T].$$

The idea is to take the limit of $\lambda \to \infty$ such that potential differences get arbitrarily large and by the above discussion, the time of existence should tend to zero. To solve the Schrödinger problem for $\lambda \to \infty$, we first determine the eigensystem of the Hamilton operator and then solve the Schrödinger equation in the eigenbasis. For this purpose, we multiply the Hamilton operator by λ^{-1} and obtain

$$\lambda^{-1} H([\lambda v], t) = V([v], t) + \lambda^{-1}(T + W), \qquad \forall t \in [0, T].$$

The spectrum of $\lambda^{-1} H([\lambda v], t)$ is just the rescaled spectrum of $H([\lambda v], t)$, i.e.

$$\sigma\left(\lambda^{-1} H([\lambda v], t)\right) = \lambda^{-1} \sigma\left(H([\lambda v], t)\right), \qquad t \in [0, T]$$

with identical eigenvectors. Therefore, we can equivalently calculate the eigensystem of the rescaled Hamilton operator and multiply the resulting spectrum by λ.
To solve for the eigensystem of $\lambda^{-1} H([\lambda v], t)$, we employ perturbation theory in λ^{-1}. We consider the scalar potential $V([v], t)$ to be the unperturbed operator and calculate perturbations to its eigensystem. Note that $V([v], t)$ is a multiplicative operator and thus diagonal in the spatial basis B^N with eigenvalues $v(\mathbf{x}, t)$ for $\mathbf{x} \in I^M$ (see Equations (3.1), (3.3)).

$$V([v], t) = v(\mathbf{x}, t) = \sum_{i=1}^{N} v(x_i, t), \qquad \forall \mathbf{x} \in I^M, \forall t \in [0, T]$$

Regardless of degeneracy in its spectrum, we can apply non-degenerate perturbation theory. To clarify this, consider an eigenvector of an arbitrary eigenspace of $V([v],t)$, i.e. $e_{\mathbf{x}} \in \mathrm{Eig}\Big(v(\mathbf{x},t)\Big)$. As $V([v],t)$ is a multiplicative operator, any state is multiplied by the sum of the scalar potentials acting on each site, meaning that any permutation of the lattice sites of $e_{\mathbf{x}}$ is also an eigenvector. In consequence, all perturbations of different eigenvectors $e_{\mathbf{x}}, e_{\mathbf{y}} \in \mathrm{Eig}\Big(v(\mathbf{z},t)\Big)$ to the same eigenstate $v(\mathbf{z},t)$ for arbitrary $\mathbf{z} \in I^M$ vanish,

$$\langle e_{\mathbf{x}}, (T+W)e_{\mathbf{y}}\rangle = 0, \qquad \forall t \in [0,T]$$

as the hopping operator T only connects states with one site difference.
Keeping this in mind, we apply non-degenerate perturbation theory to $\lambda^{-1}H([\lambda v],t)$ and multiply the resulting spectrum by λ to obtain the eigensystem of the initially stated Hamilton operator $H([\lambda v],t)$. The matrix element $\langle e_{\mathbf{x}}, We_{\mathbf{y}}\rangle$ is denoted by $W_{\mathbf{x},\mathbf{y}}$.

$$\begin{aligned}
\tilde{e}_{\mathbf{x}} &= e_{\mathbf{x}} + \lambda^{-1} \sum_{e_{\mathbf{y}} \notin \mathrm{Eig}(v(\mathbf{x},t))} \frac{\langle e_{\mathbf{y}}, (T+W)e_{\mathbf{x}}\rangle}{v(\mathbf{y},t) - v(\mathbf{x},t)} e_{\mathbf{x}} + \mathcal{O}(\lambda^{-2}) \\
\tilde{v}(\mathbf{x},t) &= \lambda v(\mathbf{x},t) + W_{x,x} + \mathcal{O}(\lambda^{-1})
\end{aligned}, \qquad \forall \mathbf{x} \in I^M, \forall t \in [0,T]$$

For $\lambda \gg 1$ and within the context of perturbation theory, we identify the Hamilton operator to be $H([\lambda v],t) = V([\tilde{v}],t)$ and solve the related Schrödinger problem $\mathcal{S}$ which yields the trajectory

$$\psi([\lambda v],t) = e^{i\int_0^t V([\tilde{v}],s)\,\mathrm{d}s}\psi_0, \qquad \forall t \in [0,T].$$

Finally, we can calculate the matrix elements of the lattice Sturm-Lioville operator and determine its rank for each point in time. Following the discussion of the last section, the rank of the lattice Sturm-Liouville operator is at most of $M-2$ if a complete matrix row is vanishing for a point in time $t_0 \in [0,T]$ (see Equation (4.21)). This means that the related state $\psi([\lambda v],t_0)$ is not an element of Ω (Definition 4.9). Therefore the time t_0 states an upper bound for the time of existence $\tau^*_{n[\lambda v]}$ (Definition 4.10), such that that we have to prove the existence of a $t_0 \in [0,T]$ and show that it turns zero for $\lambda \to \infty$. For this purpose, we consider the matrix elements of the reduced lattice Sturm-Liouville operator for a fixed site $x_0 \in I$. Inserting the trajectory into the definition of the matrix

elements (Definition 4.6) yields

$$\begin{aligned} K([\lambda v],t)_{x_0,y} &= \operatorname{Re}\xi(x_0,y)e^{i\int_0^t[\tilde{v}(x_0,s)-\tilde{v}(y,s)]\,\mathrm{d}s} \\ &= |\xi(x_0,y)|\operatorname{Re}e^{i\phi(x_0,y)+i\int_0^t[\tilde{v}(x_0,s)-\tilde{v}(y,s)]\,\mathrm{d}s} \end{aligned}, \quad \forall y \neq x_0 \in I, \forall t \in [0,T].$$

Note that wee abbreviate $2T_{x_0,y}\left(\sum_{\mathbf{x}\in I^{M-1}}\psi_0^*(x_0,\mathbf{x})\psi_0^*(y,\mathbf{x})\right)$ by $\xi(x_0,y)$ and employ the Euler representation of $\xi(x_0,y) = |\xi(x_0,y)|\operatorname{Re}e^{i\phi(x_0,y)}$ to obtain the second equality. Next, we assume the non-trivial case of $\xi(x_0,y)$ being non-zero and solve for the roots of $K[\lambda v]_{x_0,y}$. In order for $K[\lambda v]_{x_0,y}$ to vanish, the real part of the exponential has to turn zero which means that the phase of the exponential has to be an odd-numbered multiplicative of π.

$$\begin{aligned} (2n+1)\,\pi &= \int_0^t[\tilde{v}(x_0,s)-\tilde{v}(y,s)]\,\mathrm{d}s + \phi(x_0,y) \\ &= \lambda\int_0^t[v(x_0,s)-v(y,s)]\,\mathrm{d}s + t\cdot[W_{x_0,x_0}-W_{y,y}] + \phi(x_0,y) \end{aligned}, \quad \forall y \neq x_0 \in I. \tag{4.26}$$

Taking the limit $\lambda \to \infty$, $K[\lambda v]_{x_0,y}$ takes its n-th root arbitrarily fast as we assumed all components of the scalar potential pairwise differ from each other. Note, since the scalar potential v was assumed to be element of V_r, we can choose $M-1$ components of v independently. By the above equation, it is thus always possible to choose a scalar potential v such that all matrix elements vanish simultaneously. In consequence, we found an arbitrary small bound on the time of existence, rigorously explaining the before discussed result of large scalar potentials differences leading to a arbitrary small times of existence. As we cannot extend the Existence theorem (Theorem 4.11), we conclude that the uniform time of existence turns out to be zero

$$\tau_{\min} = \inf_{v\in V_r}\left\{\tau^*_{n[v]}\right\} = 0. \tag{4.27}$$

We illustrate the above discussion for an auxiliary Schrödinger problem with $N=2$ particles and $M=2$ lattice sites, i.e we consider a Schrödinger problem with the state space $\ell^2(4) \cong \mathbb{C}^4$ (see Equation (2.1.2)). We choose a common real hopping parameter $T_{x,y} = 0.5$ for all sites $x, y \in I$ and set the matrix elements of the multi-particle interaction operator W to be $w_{1,1} = w_{2,2} = 1$ and $w_{2,1} = 0$. The scalar potential

is assumed to be $v = v(1)e_1 - v(1)e_2 \in V_r$ (Definition 4.8) with the non-zero scalar potential $v(1,t) = t + 1$ for all t in $[0,T]$. Then, the resulting Hamilton operator is of the form

$$H([\lambda v],t) = 2\lambda v(1,t) \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix} + 0.5 \begin{pmatrix} 2 & 1 & 1 & 0 \\ 1 & 0 & 0 & 1 \\ 1 & 0 & 0 & 1 \\ 0 & 1 & 1 & 2 \end{pmatrix}, \quad \forall t \in [0,T]. \tag{4.28}$$

As we have chosen the same amount of lattice sites as in the previous example (see Equation ()4.24)), the lattice Sturm-Liouville operator similarly reads

$$K([\lambda v],t) = \begin{pmatrix} -K([\lambda v],t)_{1,2} & K([\lambda v],t)_{1,2} \\ K([\lambda v],t)_{1,2} & -K([\lambda v],t)_{1,2} \end{pmatrix}, \qquad \forall t \in [0,T]$$

with $K([\lambda v],t)_{1,2} = 2\,\mathrm{Re}\, T_{1,2}\langle \Pi_{1,2}\rangle_{\psi([\lambda v],t)}$. We choose the initial state to be $\psi_0 = 1/2(e_1 - e_2 - e_3 + e_4)$ which is an element of Ω because ψ_0 is the eigenstate to the lowest eigenvalue of the kinetic energy operator T (see Equation (4.15)). The resulting time evolution of the Schrödinger problem matrix element is presented in Figure 4.2.

Plot (a) presents the time evolution of the rescaled electron density for lattice site one $n([v],1)$ and the related matrix element of the lattice Sturm-Liouville operator $K[v]_{1,2}$ and the current $J[v]_{1,2}$. Compared to the non-interacting Schrödinger system presented in Figure 4.1, we find differences for all three observables within the first moments of time. This is due to the added multi-particle interaction operator W as its contribution breaks the symmetry of the scalar potential. For later times, its contribution is not of the same order as the scalar potential such that the system evolves like the non-interacting system. Note that $K[v]_{1,2}$ takes its first root at $t_0 = 1.92$, meaning that the time of existence $\tau^*_{n[v]}$ is bounded by 1.92.

We take this as a reference value and compare it to the presented roots of $K[\lambda v]_{1,2}$ for different values of $\lambda = 200, 300, 400, 500$ in plot (b). As expected, the values of the first root of $K[\lambda v]_{1,2}$ decreases with increasing values of λ; for instance, the root of $\lambda = 400$ is three orders of magnitude smaller than t_0.

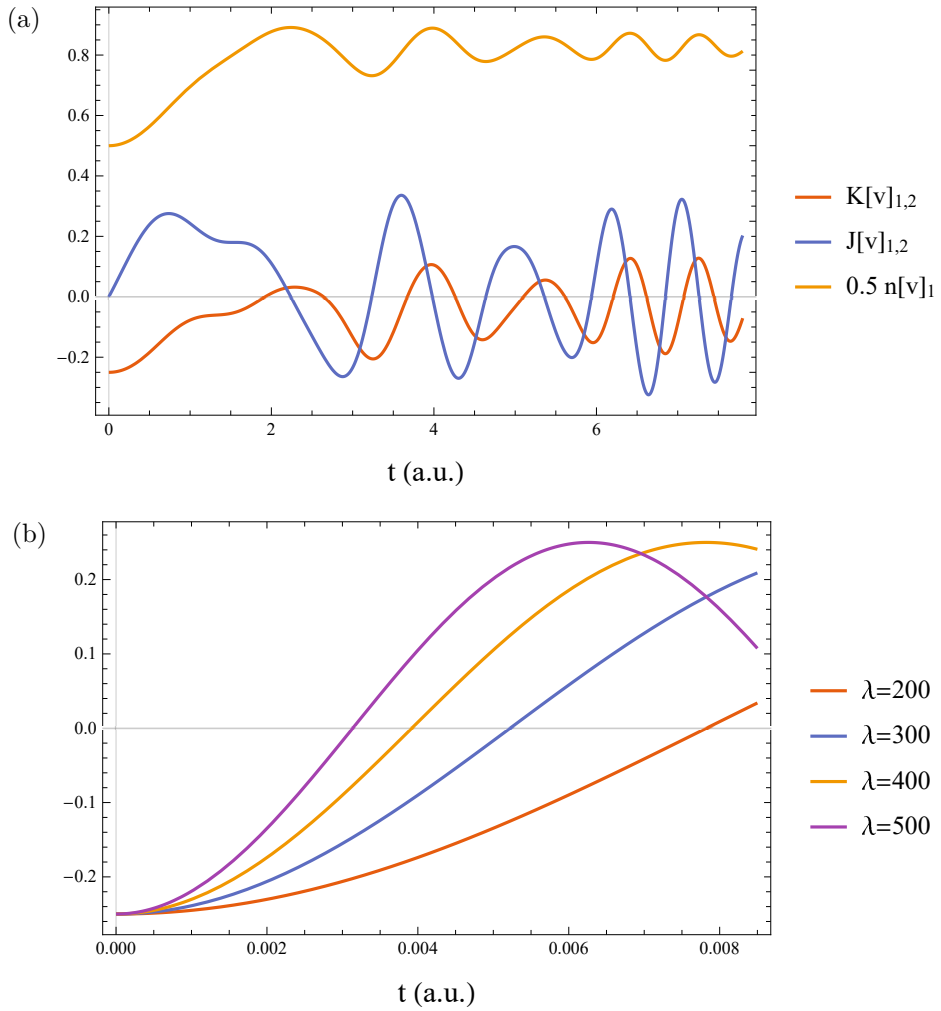

Fig. 4.2: We consider a $N = 2$ particle, $M = 2$ site Schrödinger problem with the initial state $\psi_0 = 1/2(e_1 - e_2 - e_3 + e_4)$ and Hamilton operator $H[\lambda v]$ (4.28).
(a) Time evolution of the rescaled density $0.5 \cdot n([v], 1)$ on site one and the the real and imaginary part of the complex link current $Q[v]_{1,2}$, i.e. $k[v]_{1,2}$ and $J[v]_{1,2}$ for $\lambda = 1$.
(b) Time evolution of the real part of $k[\lambda v]_{1,2}$ for different values of λ.

For $\lambda \to \infty$ the value of the first root indeed tends to zero. This result is in full agreement with the result of perturbation theory. We calculate the perturbed eigenvalues up to first order of λ^{-1},

$$-\tilde{v}(2,t) = \tilde{v}(1) = \lambda v(1,t) + 1, \qquad \forall t \in [0,T],$$

and employ Equation (4.26) to obtain the condition for the first root $\tilde{t}_{0,\lambda}$

$$\pi = \lambda \cdot \left(\tilde{t}_{0,\lambda}^2 + 2\tilde{t}_{0,\lambda}\right) + 2\tilde{t}_{0,\lambda} \tag{4.29}$$

which predicts the roots with high precision for increasing λ illustrated in Figure 4.3. It

depicts the roots $t_{0,\lambda}$ of the exact Schrödinger problem and the approximated roots $\tilde{t}_{0,\lambda}$ as a function of λ. The latter is proportional to λ^{-1} and proves high agreement with the exact roots. For instance, take $\lambda = 500$, the relative difference of the actual root and the approximated root $\tilde{t}_{0,500}$ is of order 10^{-4}. In agreement with our expectations, taking the limit $\lambda \to \infty$ yields a time of existence converging to zero.

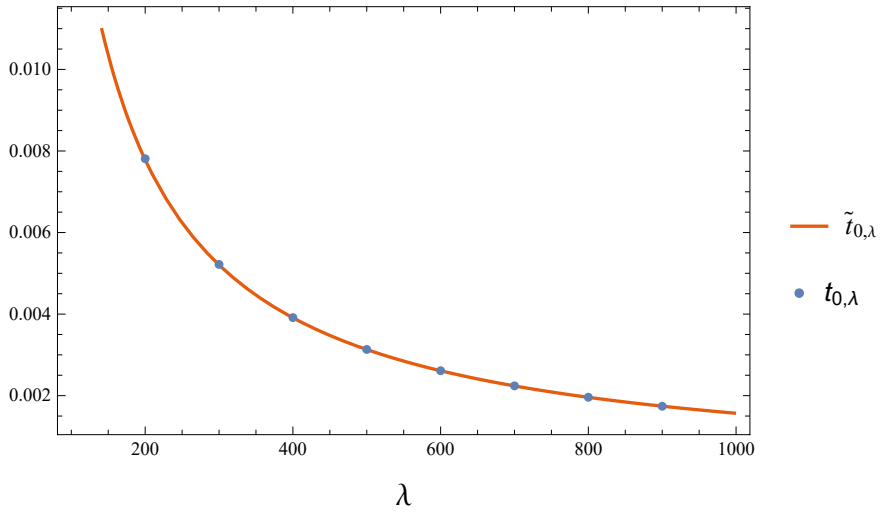

Fig. 4.3: We consider a $N = 2$ particle, $M = 2$ site Schrödinger problem with the initial state $\psi_0 = 1/2(e_1 - e_2 - e_3 + e_4)$ and Hamilton operator $H[\lambda v]$ (4.28). The approximated roots $\tilde{t}_{0,\lambda}$ of $K[v]_{1,2}$ are depicted as a function of λ (Equation (4.29)) and compared with the exact roots $t_{0,\lambda}$ of $K[v]_{1,2}$.

4.4 Potential to density diffeomorphism

Within this section, we discuss the diffeomorphic mapping property of the potential-density map $n[\cdot]$. We consider a Schrödinger problem $\mathcal{S} = \left(\psi_0, v, W\right)$ with initial state $\psi_0 \in \Omega$ (Definition 4.9) and arbitrary scalar potential $v \in V_r$ (Definition 4.8). We employ the Inverse function theorem (Theorem 2.10) and investigate the invertibility of the Fréchet derivative $D_F n[v, \cdot]$. The latter is rephrased as a linear Volterra integral equation of the second kind [22]. Invertibility is proven by applying the Banach fixed point theorem which allows for a diffeomorphic mapping property.

The Fréchet derivative $D_F n[v, \cdot]$ of the potential-density map was derived in Section 4.1. We identified $D_F n[v, \cdot]$ by a linear integral operator, describing the following linear

Volterra integral equation of first kind (4.4) [22].

$$D_F n([v,h],t) = \int_0^t k([v],t,s)h(s)\,\mathrm{d}s, \qquad \forall v,h \in V_r, \forall t \in [0,T]$$

Its integral kernel $k[v] : [0,T]^2 \to \mathcal{B}(\mathbb{R}^M)$ is twice continuously differentiable and defined by the matrix elements

$$k([v],t,s)_{x,y} = iN \sum_{j=1}^{N} \langle [\hat{\Pi}_x(t), \hat{\Pi}_y^j(s)] \rangle_{\psi_0}, \qquad \forall t \in [0,T], \forall x,y \in I.$$

with Π_x and Π_y given in the Heisenberg picture of $\mathcal{S}$ (Definition 2.7). By taking $D_F n[v,\cdot]$'s second derivative, we reformulate the Fréchet derivative as a Volterra integral equation of second kind. Note, that the integral kernel $k([v],t,s)$ vanishes for equal times $s = t$, as both projectors Π_x and Π_y^j commute for all $x, y \in I$ and $j \in \{1, ..., M\}$.

$$\partial_t^2 D_F n([v,h],t) = \partial_t k([v],t,s)\Big|_{s=t} h(t) + \int_0^t \partial_t^2 k([v],t,s)h(s)\,\mathrm{d}s, \qquad t \in [0,T]. \quad (4.30)$$

Applying the Heisenberg equation of motion enables to calculate the first derivative of the integral kernel, i.e. $\partial_t k([v],t,s)\Big|_{s=t}$. The first equality is due to the commutator $[H, \Pi_x]$ reducing to $[T, \Pi_x]$. For the second equality note that the commutator $[T, \Pi_x]$ only acts non-trivially on the first particle Hilbert space such that all $[T, \Pi_x]$ and Π_y^j commute for $j > 1$.

$$\begin{aligned} \partial_t k([v],t,s)_{x,y}\Big|_{s=t} &= -N \sum_{j=1}^{N} \langle [[T,\Pi_x],\Pi_y^j] \rangle_{\psi([v],t)} \\ &= -N \langle [[T,\Pi_x],\Pi_y] \rangle_{\psi([v],t)} \end{aligned}$$

We insert the basis representation of T and perform analogous manipulations as presented in equation (4.6) which yields the second equality.

$$\partial_t k([v],t,s)_{x,y}\Big|_{s=t} = -N \langle [[T,\Pi_x],\Pi_y] \rangle_{\psi([v],t)} = -2iN \sum_{z \in I} \langle [\mathrm{Im}\, T_{z,x} \Pi_{z,x}, \Pi_y] \rangle_{\psi([v],t)}$$

The residual commutator is rearranged, using its anti-symmetric property and the self-adjoindness of Π_x.

$$
\begin{aligned}
[\operatorname{Im} T_{z,x}\Pi_{z,x}, \Pi_y] &= \frac{[T_{z,x}\Pi_{z,x}, \Pi_y] + [\Pi_y, T_{z,x}\Pi_{z,x}]^\dagger}{2i} \\
&= -i \operatorname{Re}[T_{z,x}\Pi_{z,x}, \Pi_y]
\end{aligned}
$$

We substitute the commutator $[\operatorname{Im} T_{z,x}\Pi_{z,x}, \Pi_y]$ and identify the components of the complex link current $Q[v]$ (Definition 4.2), showing that $\partial_t k([v], t, s)\Big|_{s=t}$ equals the lattice Sturm-Liouville operator $K([v], t)$ (Definition 4.6).

$$
\begin{aligned}
\partial_t k([v], t, s)_{x,y}\Big|_{s=t} &= -2N \operatorname{Re}\left\{ \sum_{z\in I} T_{z,x} \langle [\Pi_{z,x}, \Pi_y] \rangle_{\psi([v],t)} \right\} \\
&= -2N \operatorname{Re}\left\{ \sum_{z\in I} T_{z,x} \left(\delta_{xy} \langle \Pi_{z,y} \rangle_{\psi([v],t)} - \delta_{y,z} \langle \Pi_{y,x} \rangle_{\psi([v],t)} \right) \right\} \\
&= 2 \operatorname{Re}\left\{ Q([v], t)_{x,y} - \delta_{x,y} \sum_{z\in I} Q([v], t)_{x,z} \right\} \\
&= K([v], t)_{x,y}
\end{aligned}
\tag{4.31}
$$

The equivalence of both notions is a result of $n[\cdot]$'s continuous differentiability and the twice continuous differentiability of the related physical densities $n[v]$ in time. Inserting the lattice Sturm-Liouville operator in equation (4.30) results in

$$
\partial_t^2 D_F n([v, h], t) = K([v], t) h(t) + \int_0^t \partial_t^2 k([v], t, s) h(s) \, \mathrm{d}s, \qquad t \in [0, T]. \tag{4.32}
$$

The Fréchet derivative $D_F n[v, \cdot]$ is injective if and only if equation (4.32) has a unique solution h. This is reasoned in the initial data $I(\psi_0)$ stating unique integration constants for $\partial_t^2 D_F n[v, h]$ uniquely specifying $D_F n[v, h]$. Its uniqueness can be proven by inverting the lattice Sturm-Liouville operator $K([v], t)$ in equation (4.32). It results in a fixed point type equation which allows to apply the Banach fixed point theorem.

$$
h(t) = K^{-1}([v], t) \left\{ \partial_t^2 D_F n([v, h], t) + \int_0^t \partial_t^2 k([v], t, s) h(s) \, \mathrm{d}s \right\}, \qquad \forall t \in [0, \tau^*_{n[v]}] \tag{4.33}
$$

Note, that the potential h is well-defined up to its time of existence $t \in [0, \tau^*_{n[v]}]$ because of $K([v], t)$ only being invertible within $[0, \tau^*_{n[v]}]$. This is a direct consequence of the Existence thoerem and Corollary 4.12. However, equation (4.33) being well-defined, is not a sufficient condition for the existence of an unique fixed point. To ensure the

latter, we have to guarantee that equation (4.33) defines a contraction on some Banach space. Therefore we introduce the restricted Banach space of reduced potentials $V_r|_{\tau_v}$ (Definition 4.8) with $\tau_v \in (0, \tau^*_{n[v]}]$ being the restricted time of existence. We prove that there exists some τ_v such that equation (4.33) defines a contraction G_v on $V_r|_{\tau_v}$. Note that we replaced $D_F n[v,h]$ with $D_F n \in \operatorname{Im} D_F n[v,\cdot]$.

$$\begin{aligned} G_v \;:\; V_r|_{\tau_v} &\longrightarrow V_r|_{\tau_v} \\ h &\longmapsto \Big(t \mapsto K^{-1}([v],t)\Big\{\partial_t^2 D_F n + \textstyle\int_0^t \partial_t^2 k([v],t,s)h(s)\,\mathrm{d}s\Big\}\Big) \end{aligned} \tag{4.34}$$

The map is well-defined by continuity of both functions $(t,s) \mapsto \partial_t^2 k([v],t,s)$ and $t \mapsto K^{-1}([v],t)$. The latter is due to the inverse lattice Sturm-Liouville operator being a rational function in trajectories which was proven by Farzanehpour and Tokatly (2012) [8]. If the map G_v is a contraction, invertibility of $D_F n[v,\cdot]$ on $V_r|_{\tau_v}$ is guaranteed and we can conclude a diffeomorphic mapping property of the density potential map by employing the Inverse function theorem.

Theorem 4.14 (Diffeomorphism theorem) *Let $\mathcal{S} = \big(\psi_0, v, W\big)$ be a Schrödinger problem with fixed initial state $\psi_0 \in \Omega$ and arbitrary scalar potential $v \in V_r|_{\tau_v}$ (Definition 4.8) for some* ***restricted time of existence*** *$\tau_v \in [0, \tau^*_{n[v]}]$. The potential-density map $n[\cdot]|_{U_v}$ defines a diffeomorpism on an open neighbourhood $U_v \subset V_r|_{\tau_v}$ of v if we choose the restricted time of existence to be*

$$\tau_v = \min\Big\{\tau^*_{n[v]}, \inf_{t,s\in[0,\tau^*_{n[v]}]} \Big(2 \cdot \Big\|K^{-1}([v],t)\Big\| \cdot \Big\|\partial_t^2 k([v],t,s)\Big\|\Big)^{-1}\Big\}. \tag{4.35}$$

Proof. The underlying proof idea is to employ the Inverse function theorem. The potential-density map is continuously differentiable on $V_r|_{\tau_v}$ (Definition 4.8), i.e. $n[\cdot] \in \mathcal{C}^1(V_r|_{\tau_v}, N)$ (see Section 4.1) such that we need to prove for invertibility of its Fréchet derivative $D_F n[v,\cdot]$. We thus consider the map G_v (4.34) and prove it to be a contraction for some restricted maximal time of exsitence $\tau_v \in (0, \tau^*_{n[v]}]$.

$$\begin{aligned} G_v \;:\; V_r|_{\tau_v} &\longrightarrow V_r|_{\tau_v} \\ h &\longmapsto \Big(t \mapsto K^{-1}([v],t)\Big\{\partial_t^2 D_F n + \textstyle\int_0^t \partial_t^2 k([v],t,s)h(s)\,\mathrm{d}s\Big\}\Big) \end{aligned} \tag{4.36}$$

Consider arbitrary scalar potentials $g, h \in V_r|_{\tau_v}$. We take the difference of $G_v[g]$ and

$G_v[h]$ within the V-topology (Definition 4.8) and obtain

$$\begin{aligned}\|G_v[g] - G_v[h]\|_{2,\infty} &\leq \tau_v \sup_{t,s\in[0,\tau_v]} \Big\| K^{-1}([v],t)\partial_t^2 k([v],t,s)\Big(g(s)-h(s)\Big)\Big\|_2 \\ &\leq \tau_v \sup_{t,s\in[0,\tau^*_{n[v]}]} \Big\| K^{-1}([v],t)\Big\|\Big\|\partial_t^2 k([v],t,s)\Big\|\,\|g-h\|_{2,\infty}.\end{aligned}$$

Both $t \mapsto \|K^{-1}([v],t)\|$ and $(t,s) \mapsto \|\partial_t^2 k([v],t,s)\|$ are continuous functions with a compact domain and thus take their maximum. Therefore, we can always choose a $\tau_v \in [0, \tau^*_{n[v]}]$ that satisfies

$$\tau_v = \min\Big\{\tau^*_{n[v]}, \inf_{t,s\in[0,\tau^*_{n[v]}]} \Big(2\cdot\Big\|K^{-1}([v],t)\Big\|\Big\|\partial_t^2 k([v],t,s)\Big\|\Big)^{-1}\Big\}. \tag{4.37}$$

such that G is Lipschitz and thus a contraction with $L = 1/2$. Applying the Banach fixed point theorem provides a unique fixed point of G_v, i.e. $G_v(h^*) = h^*$ for $h^* \in V_r|_{\tau_v}$ to a given $D_F n \in \operatorname{Im} D_F n[v,\cdot]$.

$$h^*(t) = K^{-1}([v],t)\Big\{\partial_t^2 D_F n(t) + \int_0^t \partial_t^2 k([v],t,s)h(s)\,\mathrm{d}s\Big\}, \qquad \forall t \in [0,\tau^*_{n[v]}] \tag{4.38}$$

We uniquely solve for $D_F n$ by employing the initial data $I(\psi_0)$ which allows to fix the integration constants. Hence, the Fréchet derivative $D_F n[v,\cdot]$ is injecitve. As $n[\cdot]|_{V_r|_{\tau_v}}$ is continuously differentiable, the inverse function theorem can be applied. It yields the existence of an open neighbourhood $U_v \subset V_r|_{\tau_v}$ of v for which $n[\cdot]|_{U_v}$ is a diffeomorphism. □

Note that the Diffeomorphism theorem imposes an additional constraint on the considered time domain of scalar potentials. Originally, a Schrödinger problem was defined on a time interval $[0,T]$, implying that scalar potentials v are continuous functions with time domain $[0,T]$, i.e. $v \in V$ (Definition 3.1). Having introduced the Existence theorem (Theorem 4.11), we had to restrict the domain of both potentials and densities to a time of existence which ensured the existence of solutions to the non-linear Schrödinger equation. In case of the Diffemorphism theorem, a diffeomorphic mapping property can only be guaranteed if we introduce another time scale, i.e. the restricted time of existence $\tau_v \in [0,\tau^*_{n[v]}]$. By now it is not clear if τ_v is non-zero for all considered scalar potential v. Hence, we investigate its relation to the set of K-invertible states in more detail (Definition 4.9) and state a lower bound on the restricted time of existence.

Corollary 4.15 *Let $\mathcal{S} = \left(\psi_0, v, W\right)$ be a Schrödinger problem with fixed initial state $\psi_0 \in \Omega$ and arbitrary scalar potential $v \in V_r$. We assume the kinetic energy operator T to be non-zero, then the restricted time of existence τ_v (see Theorem 4.14) is non-zero and bounded from below,*

$$\tau_v \geq \min\Big\{\tau^*_{n[v]}, \inf_{t\in[0,\tau^*_{n[v]}]} \|H([v],t)\|^{-1} \cdot \frac{\lambda_{\min}[v]}{4dN\|T\|}\Big\} > 0, \tag{4.39}$$

with the minimal absolute value of all possible eigenvalues of the reduced lattice Sturm-Liouville operator

$$\lambda_{\min}[v] = \inf_{t\in[0,\tau^*_{n[v]}]} \min\Big\{|\lambda| : \lambda \in \sigma\left(K_r([v],t)\right)\Big\}.$$

Proof. The restricted time of existence (4.35) is defined to be

$$\tau_v = \min\Big\{\tau^*_{n[v]}, \inf_{t,s\in[0,\tau^*_{n[v]}]} \Big(2 \cdot \Big\|K^{-1}([v],t)\Big\| \cdot \Big\|\partial_t^2 k([v],t,s)\Big\|\Big)^{-1}\Big\}. \tag{4.40}$$

As $\tau^*_{n[v]}$ is shown to be non-zero (Theorem 4.11), we only consider the non-trivial case of the restricted time of existence τ_v being smaller than $\tau^*_{n[v]}$, meaning we only have to investigate

$$\inf_{t,s\in[0,\tau^*_{n[v]}]} \Big(2 \cdot \Big\|K^{-1}([v],t)\Big\| \cdot \Big\|\partial_t^2 k([v],t,s)\Big\|\Big)^{-1} \tag{4.41}$$

to determine τ_v's lower bound. We split this question into two parts. Firstly, we estimate the operator norm of the inverse lattice Sturm-Liouville operator $K^{-1}([v],t)$ and secondly, discuss the second time derivative of the integral kernel $\partial_t^2 k([v],t,s)$ of the Fréchet derivative $D_F n[v,\cdot]$ (see Equation (4.4)).
The operator norm of the inverse lattice Sturm-Liouville operator $K^{-1}([v],t)$ is calculated to be

$$\begin{aligned}
\|K^{-1}([v],t)\| &= \sup_{y\in\mathrm{span}(\{c\})^\perp} \|K^{-1}([v],t)y\|_2 \cdot \|y\|_2^{-1} \\
&= \sup_{x\in\mathrm{span}(\{c\})^\perp, \|x\|=1} \|K([v],t)x\|_2^{-1} \\
&= \min\Big\{|\lambda| : \lambda \in \sigma\left(K_r([v],t)\right)\Big\}^{-1}
\end{aligned}$$

Firstly, we substitute the definition of the operator norm. Note that the lattice Sturm-Liouville operator is only defined for vectors $y \in \mathbb{R}^M$ being orthogonal to the space

constant vector $c = \sum_{i=1}^{M} e_i$, meaning $y \in \text{span}(\{c\})^\perp$ (see Equation (4.13), Definition 4.9). Secondly, we employ bijectivity of the lattice Sturm-Liouville operator and substitute y for $K([v],t)x$ with $x \in \{c\}^\perp$. Making use of its linearity, we rescale x to be of norm one and obtain the second equality. To obtain the last equality, we use that the lattice Sturm-Liouville operator is defined by a self-adjoint matrix and diagonalizable (Definition 4.6). Therefore, the supreme of the inverse equals the smallest norm of all eigenvalues of the spectrum of the reduced lattice Sturm-Liouville operator as we restricted x to be element of $\text{span}(\{c\})^\perp$.
Taking the infimum in time of $\|K^{-1}([v],t)\|^{-1}$ finally yields the minimal eigenvalue of the lattice Sturm-Liouville operator in time

$$\begin{aligned}\lambda_{\min}[v] &\equiv \inf_{t\in[0,\tau^*_{n[v]}]} \|K^{-1}([v],t)\|^{-1} \\ &= \inf_{t\in[0,\tau^*_{n[v]}]} \min\Big\{|\lambda| : \lambda \in \sigma\left(K_r([v],t)\right)\Big\} \\ &> 0.\end{aligned} \tag{4.42}$$

Note that $\lambda_{\min}[v]$ is non-zero as we consider the scalar potential $v \in V_r$ only up to the time of existence such that the reduced lattice Sturm-Liouville operator is of full rank, i.e. its kernel is trivial.

Next, we calculate the explicit form of the intergral kernel $\partial_t^2 k([v],t,s)$ (see Equation (4.4)) by employing the Heisenberg equation of motion twice. The operators are transformed in the Heisenberg picture of the Schrödinger problem $\mathcal{S}$ (Definition 2.7).

$$\partial_t^2 k([v],t,s)_{x,y} = N\sum_{j=1}^{N}\langle[[\hat{H}([v],t),[\hat{T}(t),\hat{\Pi}_x(t)]],\hat{\Pi}_y^j(s)]\rangle_{\psi_0}, \qquad \forall x,y \in I$$

We consider its absolute value and use the Cauchy-Schwarz inequality. We rewrite the commutators as a difference of operators and employ that the operator norm is sub-multiplicative.

$$\begin{aligned}|\partial_t^2 k([v],t,s)_{x,y}| &\leq N\sum_{j=1}^{N}\Big\|[[\hat{H}([v],t,t),[\hat{T}(t),\hat{\Pi}_x(t)]],\hat{\Pi}_y^j(s)]\Big\| \\ &\leq 8N^2\|T\|\cdot\|H([v],t)\|\end{aligned} \qquad \forall x,y \in I \tag{4.43}$$

Therefore, we estimate the operator norm of $\partial_t^2 k([v],t,s)$ by M times the largest matrix

element (4.43) such that the infimum of the inverse of the operator norm in time reads

$$\inf_{t,s\in[0,\tau^*_{n[v]}]} \left\|\partial_t^2 k([v],t,s)\right\|^{-1} \geq \inf_{t\in[0,\tau^*_{n[v]}]} \left(8dN\|T\| \cdot \|H([v],t)\|\right)^{-1} > 0. \tag{4.44}$$

Equation (4.44) is non-zero as we assume $\|T\| \neq 0$ and by continuity of the scalar potential $t \mapsto \|H([v],t)\|$, the infimum of $\|H([v],t)\|$ is bounded. Finally, we can estimate Equation (4.41) by substituting $\lambda_{\min}[v]$ (4.42) and the above equation (4.44) which yields

$$\tau_v \geq \inf_{t\in[0,\tau^*_{n[v]}]} \|H([v],t)\|^{-1} \cdot \frac{\lambda_{\min}[v]}{4dN\|T\|}$$

We obtain the desired result by inserting this lower bound into the definition of the restricted time of existence (4.35)

$$\tau_v \geq \min\left\{\tau^*_{n[v]}, \inf_{t\in[0,\tau^*_{n[v]}]} \|H([v],t)\|^{-1} \cdot \frac{\lambda_{\min}[v]}{4dN\|T\|}\right\} > 0.$$

□

By Corollary 4.15, we can always find a non-zero restricted time of existence $\tau_v \in [0, \tau^*_{n[v]}]$ such that the potential-density map defines a diffeomorphism on an open neighbourhood $U_v \subset V_r|_{\tau_v}$ of v. Note that the lower bound also varies with the scalar potential v as it again depends on the v. We investigate the dependence on the scalar potential in more detail, considering

$$\inf_{t\in[0,\tau^*_{n[\mu v]}]} \|H([\mu v],t)\|^{-1} \cdot \frac{\lambda_{\min}[v]}{4dN\|T\|} \tag{4.45}$$

for a non-zero scalar potential $v \in V_r$ scaled by the factor $\mu \in \mathbb{R}_+$. Firstly, we estimate the inverse of the operator norm of the Hamilton operator

$$\|H([\mu v],t)\| = \|T + V([\mu v],t) + W\| \leq \mu\|v\|_{2,\infty} + \|T + W\|$$

and secondly, we substitute it back into Equation (4.45) which yields

$$(4.44) \geq \inf_{t\in[0,\tau^*_{n[\mu v]}]} \left(\mu\|v\|_{2,\infty} + \|T + W\|\right)^{-1} \cdot \frac{\lambda_{\min}[\mu v]\|T\|}{4dN}$$

Note that the lattice Sturm-Liouville operator is bounded, implying its eigenvalues to be bounded. Taking the limit $\mu \to \infty$ therefore results in the above equation to converge to zero which is in full agreement with the discussion on the uniform time of

existence in Section 4.3.3.

Chapter 5

The Kohn-Sham Iteration Scheme

In the previous chapter, we established the potential-density map $n[\cdot]$ (Definition 4.1) and proved the existence of lattice TDDFT in terms of the Existence theorem. Within this chapter, we employ these results and introduce the Kohn-Sham approach towards lattice TDDFT. Using the Existence theorem, we introduce the Kohn-Sham system, i.e. a non-interacting, thus effectively single particle Schrödinger problem generating the same density as an interacting Schrödinger problem up to the common time of existence (Corollary 4.13). We establish the Kohn-Sham potential map (Definition 5.2) and the Kohn-Sham Iteration scheme (Definition 5.3), allowing to iteratively calculate the actual density of a prescribed interacting Schrödinger problem. Employing the Diffeomorphism theorem, we prove the Kohn-Sham Iteration Scheme to converge using a Banach fixed point approach and discuss emerging difficulties.

Given an arbitrary interacting Schrödinger problem, we are interested in finding its physical density. Therefore, we have to solve the related many-particle problem of N particles and M sites, meaning a system of N coupled M dimensional ordinary differential equations. With increasing particles and sites, the amount of complexity within the system increases exponentially, not allowing for accurate numerical solutions. The Existence theorem yields a conceptual solution to this problem. It allows to relate the interacting with a non-interacting Schrödinger problem, i.e. a system of N decoupled M dimensional ordinary differential equations - the Kohn-Sham system.

Definition 5.1 *Consider an interacting Schrödinger problem* $\mathcal{S}_{\text{int}} = \left(\psi_0, v_{\text{int}}, W\right)$ *with initial state* $\psi_0 \in \Omega$ *and scalar potential* $v_{\text{int}} \in V_r$ *(Definitions 4.9, 4.8). Its physical density* $n[v_{\text{int}}]$ *is denoted by* n_{int} *(Definition 4.1). The* ***Kohn-Sham system*** *is defined to be the non-interacting Schrödinger problem* $\mathcal{S}_{KS} = \left(\phi_0, v_{KS}, 0\right)$ *with initial state* $\phi_0 \in \Omega$ *and the* ***Kohn-Sham potential*** $v_{KS} \in \mathcal{V}$ *(Corollary 4.13), which generates the same density* n_{int} *as the Schrödinger problem* $\mathcal{S}_{\text{int}}$ *in a time interval* $[0, \tau] \subseteq [0, \tau^*_{v_{\text{int}}}]$. *The time* $\tau^*_{v_{\text{int}}}$ *denotes the common time of existence of* $\mathcal{S}_{\text{int}}$ *and* $\mathcal{S}_{KS}$.

Note that the Kohn-Sham system is guaranteed to exist, no matter which interacting Schrödinger problem is considered. We can always choose an initial state ϕ_0 of the Kohn-Sham system $\mathcal{S}_{\text{KS}}$ such that its initial data is identical to the initial data of the interacting Schrödinger problem $\mathcal{S}_{\text{int}}$, i.e. $I(\phi_0) = I(\psi_0)$ (Definition 4.7). Then, by Corollary 4.13, the interacting density n_{int} is v-representable (Definition 4.5) for both $\mathcal{S}_{\text{int}}$ and $\mathcal{S}_{\text{KS}}$ if we restrict its time domain to $[0, \tau^*_{v_{\text{int}}}]$. Moreover, any restriction of its time domain to $[0, \tau] \subseteq [0, \tau^*_{v_{\text{int}}}]$ is also $\mathcal{S}_{\text{int}}$ and $\mathcal{S}_{\text{KS}}$ v-representable, guaranteeing the existence of a Kohn-Sham system. We refer to v-representability of $\mathcal{S}_{\text{int}}$ and any other non-interacting Schrödinger problem by interacting and non-interacting v-representability (Definition 4.5).

Despite knowing about the existence of a Kohn-Sham system, it is not yet clear how to determine it. Simplifying the interacting Schrödinger problem to the Kohn-Sham system comes at the expense of an unknown Kohn-Sham potential non-trivially depending on the interacting density n_{int}. In practice, its dependence is unknown as we cannot employ the Existence theorem to determine the Kohn-Sham potential due to increasing complexity of the non-linear Schrödinger equation with increasing N and M (see Equation (4.12)). However to determine a Kohn-Sham system is still possible if we introduce the Kohn-Sham potential map.

The idea of the Kohn-Sham potential map is to consider a non-interacting, thus effectively single particle Schrödinger problem $\mathcal{S}_s = \left(\phi_0, v_s, 0\right)$ with arbitrary scalar potential $v_s \in V_r$ and initial state $\phi_0 \in \Omega$ with identical initial data as the prescribed interacting Schrödinger problem $\mathcal{S}_{\text{int}}$. We want to determine v_s such that it equals the Kohn-Sham potential of the interacting Schrödinger problem. Therefore, we relate both the interacting and non-interacting Schrödinger problem via their density-potential maps which allows to prove the non-interacting Schrödinger problem to be identical to

desired Kohn-Sham system for the interacting density, i.e.

$$\mathcal{S}_s = \mathcal{S}_{\mathrm{KS}}.$$

In practice, this approach is applicable as several approximations can be made. Within this thesis, we do not discuss these approximations and the reader is referred to existing literature on the topic for more details [26]. Instead, we solely consider the exact Kohn-Sham potential map and employ it for a construction of an exact Kohn-Sham Iteration scheme. Note, we first introduce an auxiliary map, the extended Kohn-Sham potential map, to define the Kohn-Sham potential map, allowing to show that its image equals the set of scalar potentials $\mathcal{V}$. The latter is shown to be necessary in order to establish the Kohn-Sham Iteration (Definition 5.3).

Definition 5.2 *Let $\mathcal{S}_{\mathrm{int}} = \left(\psi_0, v_{\mathrm{int}}, W\right)$ be an interacting Schrödinger problem with initial state $\psi_0 \in \Omega$ and scalar potential $v_{\mathrm{int}} \in V_r$. Consider a non-interacting, effective single particle Schrödinger problem $\mathcal{S}_s$ with arbitrary scalar potential $v_s \in V_r$ and initial state $\phi_0 \in \Omega$ with initial data $I(\psi_0) = I(\phi_0)$ (Definition 4.7). The potential-density maps of $\mathcal{S}_{\mathrm{int}}$ and $\mathcal{S}_s$ are denoted by $n[\cdot]$ and $n_s[\cdot]$ (Definition 4.1). The **extended Kohn-Sham potential map** $\tilde{v}_{KS}[\cdot] : \mathcal{D} \to V_r$ is defined to be*

$$\tilde{v}_{KS}([n],t) = \begin{cases} v_{\mathrm{int}}(t) - v([n],t) + v_s([n],t), & t \in [0, \tau^*_{v[n]}] \\ v_{\mathrm{int}}(t) - v([n],\tau^*_{v[n]}) + v_s([n],\tau^*_{v[n]}), & t \in (\tau^*_{v[n]}, T] \end{cases}$$

*where $\mathcal{D}$ denotes the set of interacting and non-interacting v-representable densities and $\tau^*_{v[n]}$ the common time of existence to the scalar potential $v[n]$ (see Corollary 4.13). Then, we define the **Kohn-Sham potential map** $v_{KS}[\cdot] : \mathcal{D} \to \mathcal{V}$ to be the restriction of the extended Kohn-Sham potential map to its common time of existence $\tau^*_{\tilde{v}_{KS}[n]}$.*

$$v_{KS}([n],t) = \tilde{v}_{KS}([n],t), \qquad \forall t \in [0, \tau^*_{\tilde{v}_{KS}[n]}]$$

For the extended Kohn-Sham potential map to be well-defined, we employ Corollary 4.13. It yields the invertibility of both potential-density maps $n[\cdot]$ and $n_s[\cdot]$ on the domain $\mathcal{V}$ with the interacting and non-interacting v-representable image $\mathcal{D}$. In consequence, both $v[\cdot]$ and $v_s[\cdot]$ are well-defined for densities $n \in \mathcal{D}$, returning scalar potentials $v \in \mathcal{V}$

which demands the time domain of the first sub-function to be restricted to $[0, \tau^*_{v[n]}]$. The second sub-function corresponds to its continuous continuation on the full time domain $[0, T]$ by the interacting potential v_{int}.

We employ the extended Kohn-Sham potential map to define the actual Kohn-Sham potential map. For a given density $n \in \mathcal{D}$, we define the Kohn-Sham potential map $v_{\text{KS}}[n]$ to equal $\tilde{v}_{\text{KS}}[n]$ with its time domain being restricted up to its common time of existence, i.e. $[0, \tau^*_{\tilde{v}_{\text{KS}}[n]}]$. By restricting the time domain to its own common time of existence, $v_{\text{KS}}[n]$ is an element of the set of scalar potentials $\mathcal{V}$ (see Corollary 4.13), meaning that the image of the Kohn-Sham potential map equals $\mathcal{V}$. The latter is of major importance for the Kohn-Sham Iteration and will be discussed in more detail (see Equation 5.2).

Next we want to check if the Kohn-Sham potential maps allows to determine the actual Kohn-Sham system. We therefore calculate the Kohn-Sham potential map for the interacting density n_{int}. Note, by invertibility of $v[\cdot]$ that the interacting potentials $v_{\text{int}}(t)$ and $v([n_{\text{int}}], t)$ are equivalent, meaning that we identify its common time of existence $\tau^*_{\tilde{v}_{\text{KS}}[n_{\text{int}}]}$ by $\tau^*_{v_{\text{int}}}$. As both interacting potentials are cancelling, the Kohn-Sham potential map reads

$$v_{\text{KS}}([n_{\text{int}}], t) = v_s([n_{\text{int}}], t), \qquad \forall t \in [0, \tau^*_{v_{\text{int}}}].$$

It remains to prove the corresponding non-interacting Schrödinger problem $\mathcal{S}_s = \left(\phi_0, v_{\text{KS}}[n_{\text{int}}], 0\right)$ to be the Kohn-Sham system, requiring the density to reproduce the prescribed density n_{int} in a time interval $[0, \tau] \subseteq [0, \tau^*_{v_{\text{int}}}]$. We solve for the density by applying the density-potential map $n_s[\cdot]$ and employ invertibility of $n_s[\cdot]$ on its domain $\mathcal{D}$.

$$n_s\left(v_s([n_{\text{int}}], t)\right) = n_{\text{int}}(t), \qquad \forall t \in [0, \tau^*_{v_{\text{int}}}]$$

As the Schrödinger problem $\mathcal{S} = \left(\phi_0, v_{\text{KS}}[n_{\text{int}}], 0\right)$ is shown to reproduce the prescribed density n_{int} within the time interval $[0, \tau^*_{v_{\text{int}}}]$, we identify $v_{\text{KS}}[n_{\text{int}}]$ with the Kohn-Sham potential v_{KS} and $\mathcal{S}_s$ with the Kohn-Sham system $\mathcal{S}_{\text{KS}}$.

We consider the Kohn-Sham potential to be the starting point for further investigations of a Kohn-Sham system. Following the above calculation, we note that the construction of the Kohn-Sham potential map relies on the difference of the actual pre-

scriped v_{int} and the calculated density-potential map $v[\cdot]$ of the interacting Schrödinger problem to a given density $n \in \mathcal{D}$.

$$v_{\text{int}}(t) - v([n], t), \qquad \forall t \in [0, \tau^*_{v[n]}] \tag{5.1}$$

If the difference vanishes, the non-interacting Schrödinger problem $\mathcal{S}_s = \left(\phi_0, v_{\text{KS}}[n_{\text{int}}], 0\right)$ equals the desired Kohn-Sham system, which is satisfied for $n = n_{\text{int}}$. By continuity of the potential-density map (see Diffeomorphism Theorem), we expect the difference in Equation (5.1) to be small for densities only slightly differing from the actual interacting density. Therefore, decreasing difference in densities might imply convergence of the non-interacting Schrödinger problem to the actual Kohn-Sham system. We follow this approach, trying to establish a contractive iteration scheme - the Kohn-Sham Iteration scheme. We first present its general idea, safely skipping mathematical details.
As a starting point, we consider the prescriped interacting Schrödinger problem $\mathcal{S}_{\text{int}} = \left(\psi_0, v_{\text{int}}, W\right)$ and non-interacting Schrödinger problem $\mathcal{S}_s$ with initial state ϕ_0 and initial data $I(\phi_0) = I(\psi_0)$. Firstly, we choose an arbitrary density $n_0 \in \mathcal{D}$ and calculate the related scalar potentials of the non-interacting Schrödinger and interacting Schrödinger problem, i.e. $v_s[n_0]$ and $v[n_0]$. Secondly, we insert the results into the Kohn-Sham potential map, solving the non-interacting Schrödinger problem $\left(\phi_0, v_{\text{KS}}[n_0], 0\right)$ for the corresponding non-interacting density denoted by n_1. It is important to note that the image of the Kohn-Sham potential map is identical to $\mathcal{V}$ (Definition 5.2). Therefore, the calculated non-interacting density is again interacting and non-interacting v-representable, analogous to the density n_0.

$$n_1 = n_s \circ v_{\text{KS}}[n_0] \in \mathcal{D} \tag{5.2}$$

Being an element of $\mathcal{D}$, we can apply the Kohn-Sham potential map to n_1 and repeat the exact same procedure as before to obtain n_2 and so forth, defining an iterative sequence of densities - the Kohn-Sham Iteration which is illustrated in Figure 5.1.

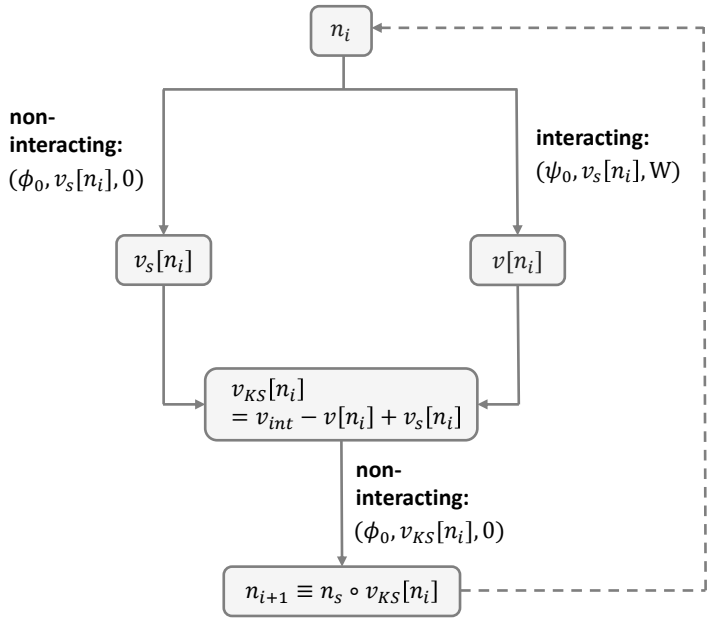

Fig. 5.1: The Kohn-Sham Iteration scheme for a prescribed interacting Schrödinger problem $\mathcal{S}_{\text{int}} = \left(\psi_0, v_{\text{int}}, W\right)$ with initial state $\psi_0 \in \Omega$ and scalar potential $v_{\text{int}} \in V_r$. We consider the non-interacting Schrödinger problem $\mathcal{S}_s$ with arbitrary scalar potential $v_s \in V_r$ and initial state $\phi_0 \in \Omega$ with initial data $I(\phi_0) = I(\psi_0)$. The initial density n_0 is assumed to be arbitrary in $\mathcal{D}$.

Within the following section, we want to investigate the possibility of convergence of the Kohn-Sham Iteration to the interacting density of $\mathcal{S}_{\text{int}}$. This requires a more rigorous mathematical treatment and an exact definition of the previous described Kohn-Sham Iteration scheme.

Definition 5.3 (Kohn-Sham Iteration) *Let $\mathcal{S}_{\text{int}} = \left(\psi_0, v_{\text{int}}, W\right)$ be an interacting Schrödinger problem with initial state $\psi_0 \in \Omega$ and scalar potential $v_{\text{int}} \in V_r$. Consider a non-interacting Schrödinger problem $\mathcal{S}_s$ with arbitrary scalar potential $v_S \in V_r$ and initial state $\phi_0 \in \Omega$ with initial data $I(\psi_0) = I(\phi_0)$. We define the map*

$$\begin{aligned} \Phi \;:\; \mathcal{D} &\longrightarrow \mathcal{D} \\ n &\longmapsto \Phi[n] = n_s \circ v_{KS}[n] \end{aligned}$$

*where $v_{KS}[n]$ denotes the Kohn-Sham potential map (Definition 5.2). The map Φ defines the **Kohn-Sham Iteration scheme** which corresponds to the recursively defined sequence $(n_k)_{k\in\mathbf{N}}$ with elements $n_{k+1} \equiv \Phi[n_k]$ for arbitrary initial values $n_0 \in \mathcal{D}$.*

Following its definition, the Kohn-Sham Iteration scheme equals exactly the definition of the Banach Iteration Scheme (see Theorem 2.2). Hence, we can investigate its convergence applying the Banach fixed point theorem and prove for Φ to admit a unique fixed point. For now, we assume that Φ admits a unique fixed point, i.e. $n^* \in \mathcal{D}$ such that $\Phi[n^*] = n^*$. Then, the fixed point can be shown to equal the density of the interacting Schrödinger problem within a non-zero time domain. Without loss of generality, we assume the common time of existence $\tau^*_{\tilde{v}_{\mathrm{KS}}[n^*]}$ to be bounded by $\tau^*_{v[n^*]}$, meaning

$$v_{\mathrm{KS}}([n^*], t) = v_{\mathrm{int}}(t) - v([n^*], t) + v_s([n^*], t), \qquad \forall t \in [0, \tau^*_{\tilde{v}_{\mathrm{KS}}[n^*]}].$$

Employing the fixed point property yields

$$\begin{aligned} v_{\mathrm{KS}}([n^*], t) &= v_{\mathrm{int}}(t) - v([n^*], t) + v_s \circ \Phi[n^*](t) \\ &= v_{\mathrm{int}}(t) - v([n^*], t) + v_s \circ n_s \circ v_{\mathrm{KS}}[n^*](t), \qquad \forall t \in [0, \tau^*_{\tilde{v}_{\mathrm{KS}}[n^*]}] \\ &= v_{\mathrm{int}}(t) - v([n^*], t) + v_{\mathrm{KS}}([n^*], t) \end{aligned}$$

which proves the equivalence of the interacting potential v_{int} and the calculated interacting density-potential map $v[n^*]$ within the time interval $[0, \tau^*_{\tilde{v}_{\mathrm{KS}}[n^*]}]$. Applying the potential-density map produces the desired result, i.e.

$$n_{\mathrm{int}}(t) = n^*(t), \qquad \forall t \in [0, \tau^*_{\tilde{v}_{\mathrm{KS}}[n^*]}]. \tag{5.3}$$

Hence, instead of proving convergence to the density of the interacting Schrödinger problem, we can prove the existence of a unique fixed point of the map Φ.

5.1 A Banach fixed point approach towards convergence

Within this section, we present a sufficient condition for the convergence of the Kohn-Sham Iteration scheme to the density of a prescribed interacting Schrödinger problem (Definition 5.3). It demands the initial density to be chosen such that it is contained within a compact neighbourhood $\mathcal{N}$ of the interacting density on which the map Φ defines a diffeomorphism.

To prove the Kohn-Sham Iteration Scheme to converge, we follow the discussion of the last Section and investigate the map Φ to admit a unique fixed point employing the Banach fixed point theorem (Theorem 2.2). The main idea is to utilize the results about the density-potential map discussed in Chapter 4. Employing the Diffeomorphism Theorem (Theorem 4.14), we prove Φ to define a diffeomorphism on a proper compact subset of the Banach space

$$\mathcal{N} \subset \Big(\mathcal{C}([0,\tau],\mathbb{R}^M), \|\cdot\|_{2,\infty}\Big)$$

for some non-zero time $\tau \in (0,T]$. Next, we employ both the diffeomorphic mapping property and pre-compactness of physical densities (Corollary 4.4) to establish a contractive mapping, i.e. there exists a $L \in [0,1)$ such that

$$\|\Phi[n] - \Phi[\tilde{n}]\|_{2,\infty} \leq L\|n - \tilde{n}\|_{2,\infty}, \quad \forall n, \tilde{n} \in \mathcal{N}$$

Then, applying the Banach fixed-point theorem yields the desired result of the existence of a unique fixed point. This approach faces the issue that the existence of a unique fixed-point depends on the chosen initial density n_0 which we are going to discuss in more detail.

Theorem 5.4 (Kohn-Sham theorem) *Consider the interacting Schrödinger problem $\mathcal{S}_{\text{int}}$ with scalar potential $v_{\text{int}} \in V_r$ (Definition 4.8) and initial state $\psi_0 \in \Omega$ (Definition 4.9). There exists a non-trivial time domain $[0,\tau] \subseteq [0,T]$ of the interacting density n_{int} for which Φ (Definition 5.3) defines a diffeomorphism on a compact convex neighbourhood $\mathcal{N}$ of n_{int},*

$$\mathcal{N} \subset \Big(C([0,\tau],\mathbb{R}^M), \|\cdot\|_{2,\infty}\Big),$$

ensuring convergence of the Kohn-Sham Iteration Scheme for any initial density $n_0 \in \mathcal{N}$.

Proof. Throughout this proof, the non-interacting Schrödinger problem is chosen to be $S = \Big(\phi_0, v, 0\Big)$ with arbitrary scalar potential $v \in V_r$ (Definition 4.8) and initial state $\phi_0 \in \Omega$ with $I(\psi_0) = I(\phi_0)$ (Definition 4.7). We first consider the interacting and non-interacting potential-density maps $n[\cdot]$ and $n_s[\cdot]$. Employing the Diffeomorphism theorem (Theorem 4.14), both $n[\cdot]$ and $n_s[\cdot]$ define a diffeomorphism on some open

neighbourhood

$$U_{v_{\text{int}}} \subset \left(C([0,\tau],\mathbb{R}^M), \|\cdot\|_{2,\infty}\right)$$

for some non-zero time τ. It is bounded by the minimum of $n[\cdot]$'s and $n_s[\cdot]$'s restricted times of existence and the common time of existence of $\mathcal{S}_{\text{int}}$ and $\mathcal{S} = \left(\phi_0, v_{\text{int}}, 0\right)$.

$$\tau \leq \min\{\tau_{v_{\text{int}}}, \tilde{\tau}_{v_{\text{int}}}, \tau^*_{v_{\text{int}}}\}$$

Choosing the minimum of both restricted times of existence ensures both $n[\cdot]$ and $n_s[\cdot]$ to define diffeomorphisms on $U_{v_{\text{int}}}$ (see Diffeomorphism theorem). Also including the common time of existence $\tau^*_{v_{\text{int}}}$ allows to employ Corollary 4.13 such that both $n[\cdot]$ and $n_s[\cdot]$ generate the exact same image, i.e. identical sets of physical densities

$$n[U_{v_{\text{int}}}] = n_s[U_{v_{\text{int}}}] \subset \left(C([0,\tau],\mathbb{R}^M), \|\cdot\|_{2,\infty}\right). \tag{5.4}$$

Note that Φ is defined to be the composition of potential-density maps such that it also defines a diffeomorphism on any subset of $n[U_{v_{\text{int}}}]$. We wisely choose some convex closed subset $\mathcal{N} \subset n[U_{v_{\text{int}}}]$.

$$\begin{aligned} \Phi \;:\; \mathcal{N} &\longrightarrow \mathcal{N} \\ n &\longmapsto n_s \circ v_{\text{KS}}[n] \end{aligned}$$

As we defined the set of physical densities $n[U_{v_{\text{int}}}]$ to be a subset of the continuous function space $C([0,\tau],\mathbb{R}^M)$ equipped with the supremum norm, we can employ Corollary 4.4. It states the pre-compactness of the set of physical densities $n[U_{v_{\text{int}}}]$, implying compactness of the closed subset $\mathcal{N}$.

It is left to prove that Φ defines a contraction. For this purpose, we consider the difference of two arbitrary densities $n, \tilde{n} \in \mathcal{N}$. As Φ defines a diffeomorphism, we can employ the fundamental theorem of calculus and rewrite the difference using the Fréchet derivative of Φ. The convex combination of n and $\tilde{n}$ is denoted by $n_\lambda \equiv \tilde{n} + \lambda(n - \tilde{n})$.

$$\Phi[n] - \Phi[\tilde{n}] = \int_0^1 D_F\Phi[n_\lambda, n - \tilde{n}]d\lambda$$

Next, we apply the $\|\cdot\|_{2,\infty}$-norm. Firstly, we employ linearity of the Fréchet derivative. Secondly, we estimate the integral, taking the supremum over all considered densities

$\mathcal{N}$.

$$\begin{aligned} \|\Phi[n] - \Phi[\tilde{n}]\|_{2,\infty} &\leq \|n - \tilde{n}\|_{2,\infty} \int_0^1 \|D_F \Phi[n_\lambda]\| \, \mathrm{d}\lambda \\ &\leq \|n - \tilde{n}\|_{2,\infty} \cdot \sup_{n \in \mathcal{N}} \|D_F \Phi[n]\| \end{aligned} \tag{5.5}$$

Identifying $\sup_{n\in\mathcal{N}} \|D_F\Phi[n]\|$ with the desired Lipschitz-constant L, it only remains to prove it to be smaller than one. Therefore, we investigate the Fréchet derivative in more detail. We first note that Φ is a composite function of $n_s[\cdot]$ and $v_{\mathrm{KS}}[\cdot]$ allowing to estimate its Fréchet derivative applying the chain rule. Next, we insert the definition of the Fréchet derivative of the potential-density map $n_s[\cdot]$ (see Equation (4.4)). Taking the supremum in time of its operator norm yields the second inequality.

$$\begin{aligned} \sup_{n\in\mathcal{N}} \|D_F\Phi[n]\| &\leq \sup_{n\in\mathcal{N}} \|(D_F n_s)[v_{\mathrm{KS}}[n]]\| \cdot \|D_F v_{\mathrm{KS}}[n]\| \\ &\leq \tau \cdot \sup_{n\in\mathcal{N}} \|k[v_{\mathrm{KS}}[n]]\| \cdot \|D_F v_{\mathrm{KS}}[n]\| \end{aligned} \tag{5.6}$$

Both $n \mapsto \|k[v_{\mathrm{KS}}[n]]\|$ and $n \mapsto \|D_F v_{\mathrm{KS}[n]}\|$ are continuous functions on a compact domain $\mathcal{N}$ and thus the supremum is a maximum. In consequence, we can always assume the non-zero time $\tau \leq \min\{\tau_{v_{\mathrm{int}}}, \tilde{\tau}_{v_{\mathrm{int}}}, \tau^*_{v_{\mathrm{int}}}\}$ to be choosen such that Equation (5.6) is bounded by $1/2$. Substituting this estimate into Equation (5.5) yields the desired contraction propperty of Φ with contraction constant $L = 1/2$.

$$\|\Phi[n] - \Phi[\tilde{n}]\|_{2,\infty} \leq 1/2 \|n - \tilde{n}\|_{2,\infty}, \qquad \forall n, \tilde{n} \in \mathcal{N}$$

We apply the Banach fixed point theorem (Theorem 2.2) which proves the existence of a unique fixed point $n^* \in \mathcal{N}$ of Φ, i.e. $\Phi[n^*] = n^*$ and we conclude that the Kohn-Sham Iteration Scheme converges to the interacting density if we assume the initial density to be element of the compact subset $\mathcal{N}$. □

This result is of major importance as it provides a rigorous formulation of a Kohn-Sham approach towards TDDFT. We want to conclude with some final remarks on the introduced set $\mathcal{N}$.

5.2 Concluding remarks: The time of existence revisited

> That doesn't mean anything. Maybe it was yesterday.
>
> The Stranger, Albert Camus

According to the Kohn-Sham theorem (Theorem 5.4), the Kohn-Sham Sham Iteration scheme (Definition 5.3) converges to the density of the prescribed interacting Schrödinger problem if the initial density n_0 is chosen to be in $\mathcal{N}$.
However, for initial densities $n_0 \in \mathcal{D}\backslash\mathcal{N}$, convergence to the interacting density is not quite clear yet. Even if the related Kohn-Sham Iteration scheme converges in some cases, we cannot guarantee for the limit point to be the interacting density as Φ might not define a contraction on the whole set $\mathcal{D}$ (Definition 5.3). Therefore, it is worth giving a rough estimate on the size of $\mathcal{N} \subset n[U_{\text{int}}] \subset C([0,T],\mathbb{R}^M)$, assessing the likelyhood of the initial density n_0 to be chosen in practice such that $n_0 \in \mathcal{N}$.
For simplicity of the discussion, we choose $\mathcal{N}$ to be - without loss of generality - contained in an open ball $B_r[n_{\text{int}}]$ of the neighbourhood $n[U_{v_{\text{int}}}]$ with radius r and center point n_{int}. Requiring the initial density $n_0 \in \mathcal{N}$ thus equivalently reads

$$n_0 \in \mathcal{N} \subset B_r[n_{\text{int}}] \equiv \Big\{ n \in C([0,\tau],\mathbb{R}^M) \;\Big|\; \|n - n_{\text{int}}\|_{2,\infty} \leq r \Big\},$$

meaning that the difference of n_0 and n_{int} in the $\|\cdot\|_{2,\infty}$-norm is limited by the radius r. Then, following the Diffeomorphism theorem (Theorem 4.14), the size of the ball $B_r[n_{\text{int}}]$ is determined using the Inverse function theorem (Theorem 2.10). By the Inverse function theorem, we expect the radius r to be large for small values of the Fréchet derivative $\|D_F\Phi[n]\|$ around n_{int} and vice versa. From a physical point of view, we therefore expect the sufficient condition of $n_0 \in \mathcal{N}$ to be only quite restrictive for strongly changing potential-density landscapes as the radius of $B_r[n_{\text{int}}]$ might be small. Then, it is not unlikely that in practice the chosen initial density n_0 is chosen such that $n_0 \in \mathcal{D}\backslash\mathcal{N}$.

This is a fundamental problem within the here taken approach of establishing Kohn-Sham TDDFT for lattice systems by inverting the lattice Sturm-Liouville operator in the force balance equation (Definition 4.6, Equation (4.10)). It necessitates to restrict the time domain of both potentials and densities to their time of existence (Definition 4.10). In turn, it is not possible to establish a common non-zero time-domain as we find the uniform time of existence to be zero (see Equation (4.27)). Provided the existence of a common non-zero time domain which is right-bounded by the infimum of all possible restricted times of existence (Theorem 4.14), the potential-density map would define a diffeomorphism on $V|_r$ (Definition 4.8) employing both the Diffeomorphism and Existence theorem (Theorem 4.11). This enables to choose the compact set $\mathcal{N}$ to be equivalent to the set of physical densities $\mathcal{N} = n[V|_r]$ (Definition 4.1). In consequence, any initial density n_0 would serve for a convergent Kohn-Sham Iteration scheme.
For future investigations, it would therefore be fruitful to investigate the following: first, the initial dependence of the convergence of the Kohn-Sham Iteration scheme; but also, second, the possibility of establishing Kohn-Sham TDDFT for lattice systems, using approaches not involving explicit inversion of the lattice Sturm-Liouville operator as done by van Leeuwen [27].

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