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Numerical Methods for Wave Propagation

Analysis and Applications in Quantum Dynamics

EMIL KIERI



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Abstract

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We study numerical methods for time-dependent partial differential equations describing wave propagation, primarily applied to problems in quantum dynamics governed by the time-dependent Schrödinger equation (TDSE). We consider both methods for spatial approximation and for time stepping. In most settings, numerical solution of the TDSE is more challenging than solving a hyperbolic wave equation. This is mainly because the dispersion relation of the TDSE makes it very sensitive to dispersion error, and infers a stringent time step restriction for standard explicit time stepping schemes. The TDSE is also often posed in high dimensions, where standard methods are intractable.

The sensitivity to dispersion error makes spectral methods advantageous for the TDSE. We use spectral or pseudospectral methods in all except one of the included papers. In Paper III we improve and analyse the accuracy of the Fourier pseudospectral method applied to a problem with limited regularity, and in Paper V we construct a matrix-free spectral method for problems with non-trivial boundary conditions. Due to its stiffness, the TDSE is most often solved using exponential time integration. In this thesis we use exponential operator splitting and Krylov subspace methods. We rigorously prove convergence for force-gradient operator splitting methods in Paper IV. One way of making high-dimensional problems computationally tractable is low-rank approximation. In Paper VI we prove that a splitting method for dynamical low-rank approximation is robust to singular values in the approximation approaching zero, a situation which is difficult to handle since it implies strong curvature of the approximation space.

Keywords: computational wave propagation, quantum dynamics, time-dependent Schrödinger equation, spectral methods, Gaussian beams, splitting methods, low-rank approximation

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List of papers

This thesis is based on the following papers, which are referred to in the text by their Roman numerals.

- I E. Kieri, S. Holmgren and H. O. Karlsson. An adaptive pseudospectral method for wave packet dynamics. *J. Chem. Phys.*, 137:044111, 2012.
- II E. Kieri, G. Kreiss, and O. Runborg. Coupling of Gaussian beam and finite difference solvers for semiclassical Schrödinger equations. *Adv. Appl. Math. Mech.*, 7:687–714, 2015.¹
- III E. Kieri. Accelerated convergence for Schrödinger equations with non-smooth potentials. *BIT*, 54:729–748, 2014.
- IV E. Kieri. Stiff convergence of force-gradient operator splitting methods. *Appl. Numer. Math.*, 94:33–45, 2015.
- V B. Brumm and E. Kieri. A matrix-free Legendre spectral method for initial-boundary value problems. *Preprint, Mathematisches Institut, Universität Tübingen*, 2015. (Submitted)
- VI E. Kieri, Ch. Lubich, and H. Walach. Discretized dynamical low-rank approximation in the presence of small singular values. *Preprint, Mathematisches Institut, Universität Tübingen*, 2015. (Submitted)

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1. Introduction

With our eyes and ears, we sense optical and acoustic waves. The physics of wave propagation thereby has a strong effect on how we experience the world around us. The sound of thunder comes after the flash of lightning as sound waves travel slower than light waves, and were it not for atmospheric refraction the days would have appeared shorter. Waves are also frequently used in technology. Broadcasting and mobile telecommunication transport information using electromagnetic waves, which are the same kind of waves as visible light, only with much longer wave lengths. In maritime navigation sea depth is measured by emitting sound downwards and measuring the time it takes for the echo to return. The same technique is used by bats to locate flying insects. Radar also uses echoes, of electromagnetic waves, to locate terrain obstacles and vehicles.

Optical and acoustic waves, as well as the vibrations of strings and membranes, are described by the wave equation,

$$u_{tt} = c^2 \Delta u, \quad x \in \Omega, t > 0. \quad (1.1)$$

Here, the wave speed $c = c(x) > 0$, Δ denotes the Laplace operator, and Ω is the computational domain. The wave equation is a linear hyperbolic partial differential equation (PDE), of second order in both space and time. For constant wave speed it can be solved in a few simple geometries using separation of variables [26], and on the real line there exists a simple general solution. In more general geometries or with spatially variable wave speed, numerical methods are necessary for solving (1.1).

Other types of wave phenomena satisfy different wave equations, unified by the property that they describe transport of energy via the propagation of oscillations. Seismic events trigger mechanical waves in the crust of the earth, i.e., earthquakes, which are governed by the elastic wave equation. The nonlinear Korteweg–de Vries equation can be used to describe waves in shallow waters. In this thesis we are mainly concerned with the time-dependent Schrödinger equation (TDSE) [78], which describes the time evolution of the wave function in quantum mechanics. A key concept in quantum mechanics is the wave–particle duality: Not only do we experience the world around us through waves, the world in itself consists of waves, as the state of a particle has a probabilistic description through the wave function. The wave function gives probability densities for the position, momentum, and other observables of the particle. This probabilistic nature of matter has nothing to do with our

limited capabilities for measurement, it is physically impossible to simultaneously know the position and momentum of a particle exactly. This is known as the Heisenberg uncertainty principle [37].

The Schrödinger equation reads

$$iu_t = -\frac{1}{2m}\Delta u + Vu, \quad x \in \mathbb{R}^d, t > 0, \quad (1.2)$$

where $u = u(x, t)$ is the wave function, m is the mass of the particles under consideration, and V is the potential. Since V can be an arbitrary function of x this is a variable coefficient problem where analytical solutions are known only in a small number of settings, such as the hydrogen atom [76] and the harmonic oscillator [77]. Additionally, the Schrödinger equation is more difficult than (1.1) to solve numerically. This is due to its dispersive nature, the speed of propagation is not bounded for (1.2). We will discuss this, and the implications of it, in more detail in Chapter 2.

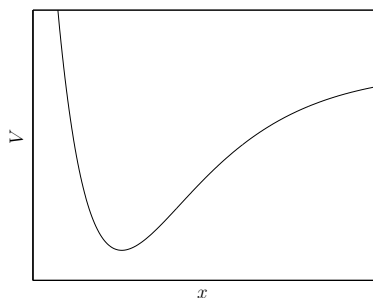


Figure 1.1. A typical example of a potential for a diatomic molecule, as function of the nuclear separation x .

An application of the Schrödinger equation which we frequently consider in this work is molecular dynamics, the study of the motion of atomic nuclei. We will typically do this under the Born–Oppenheimer approximation [8], assuming that the potential V is given. Intuitively speaking, the Born–Oppenheimer approximation says that since the nuclei are much heavier than the electrons, they move much slower. Therefore, the nuclei only sense the time-averaged distribution of electrons, which can be modelled in the potential V . The art of determining the potential is known as electronic structure theory or quantum chemistry, and features, e.g., the density functional theory [44, 50] and Hartree–Fock [84] classes of methods. Figure 1.1 illustrates what the potential might look like in a diatomic molecule, one of the simplest cases imaginable. The nuclear dynamics of a diatomic molecule can be reduced to a one-dimensional problem, with the distance between the two nuclei as spatial coordinate, since the absolute position and orientation in space of the molecule usually is of limited interest. It is possible to reduce the dimensionality also

for larger systems by eliminating external degrees of freedom and take advantage of symmetries in the system. One can, e.g, remove the absolute position (three coordinates) and orientation (three coordinates) of the system, so that a system with N nuclei yields a problem in $d = 3N - 6$ dimensions. We call the reduced configuration space internal coordinates.

In classical mechanics, one models the dynamics of the nuclei using Hamilton's equations of motion [35]. This is a much simpler model than the TDSE. With q and p denoting the positions and momenta of the nuclei, possibly in internal coordinates, Hamilton's equations read

$$\begin{aligned} \frac{dq}{dt} &= \frac{\partial H}{\partial p}, & q, p &\in \mathbb{R}^d, \\ \frac{dp}{dt} &= -\frac{\partial H}{\partial q}. \end{aligned} \tag{1.3}$$

In Cartesian coordinates, the Hamiltonian function is

$$H(q, p) = \frac{|p|^2}{2m} + V(q),$$

where the terms in the right-hand side are the kinetic and potential energies, respectively. Coordinate transformations, e.g., to internal coordinates, modify the Hamiltonian function. Eq. (1.3) is a Hamiltonian system of ordinary differential equations (ODEs). Its numerical solution is well studied, see, e.g., [32, 57], and computations with millions of nuclei are made routinely. However, not all the physics of molecular dynamics can be explained by classical mechanics, which does not take phenomena like the uncertainty principle, tunnelling or zero-point energy into account. Chemical reactions are particularly difficult to model classically. This motivates quantum mechanics, modelled by the Schrödinger equation (1.2), which offers a more accurate description, especially for light particles. As m grows, quantum dynamics approaches classical dynamics. Unfortunately, as the Schrödinger equation is a PDE, solving it is much more demanding than solving the system of ODEs (1.3). Direct solution is only possible for very small systems. The necessary amount of computational work and memory grows exponentially with the dimension, a property known as the curse of dimensionality. Direct numerical simulation becomes intractable already in five or six dimensions. Possible remedies to this will be discussed in Chapter 3. The numerical solution of the Schrödinger equation need not be easy in small dimensions either, as will be discussed in the next chapter.

2. What's difficult about computational wave propagation?

In this chapter we discuss some of the difficulties with computational wave propagation in general, and with numerical solution of the Schrödinger equation in particular.

2.1 Dispersion error

In wave propagation, errors tend to accumulate over time. This stands in contrast to parabolic problems, where small disturbances normally are smoothed out. The effect of this error accumulation is enhanced by the tendency of waves to travel over large distances. The considered domain is often many times larger than the wave length. When wave propagation problems are solved with difference methods, the type of error which most often dominates is called dispersion error, i.e., that waves are propagated with the wrong speed. This type of error is smaller if higher order discretisations are used. We illustrate this below using a simple model problem. A more detailed discussion is found in, e.g., [24].

The advection equation,

$$v_t + v_x = 0, \quad x \in \mathbb{R}, t > 0, \quad (2.1)$$

is arguably the simplest possible model problem for wave propagation. The plane waves

$$v(x, t) = e^{i\omega(x-t)}, \quad \omega \in \mathbb{R},$$

are solutions to (2.1). The waves travel with unit speed for all choices of ω . We introduce a computational grid with uniform spacing h , $x_j = jh$, $j \in \mathbb{Z}$, and discretise the spatial derivative with p th order central finite differences. This yields an infinite system of ODEs, solved by the plane waves

$$v_j(t) = e^{i\omega(x_j - c_p(\omega h)t)}, \quad j \in \mathbb{Z}, |\omega| \leq \frac{\pi}{h},$$

where $v_j(t)$ is an approximation of $v(x_j, t)$. The finite difference approximation of the spatial derivative results in a new wave speed, c_p , which now is a function of ωh . c_p is approximately 1 for small frequencies but deteriorates as $|\omega h|$ approaches π . The wave speed deteriorates slower for higher order

methods, as illustrated in Figure 2.1 (left panel). The frequency $\omega = \pi/h$ corresponds to two grid points per wave length, and is the highest representable frequency on the grid. If we demand that the pointwise error should not exceed ε after the time t , we need at least

$$M_p = C_p \left(\frac{\omega t}{\varepsilon} \right)^{1/p}$$

grid points per wave length [25, Ch. 3]. The constant C_p depends on p , but not on ω , t or ε . This expression shows that we need fewer grid points for the same accuracy if we use higher order methods, and that the benefit of using higher order methods grows if the solution is to be transported many wave lengths.

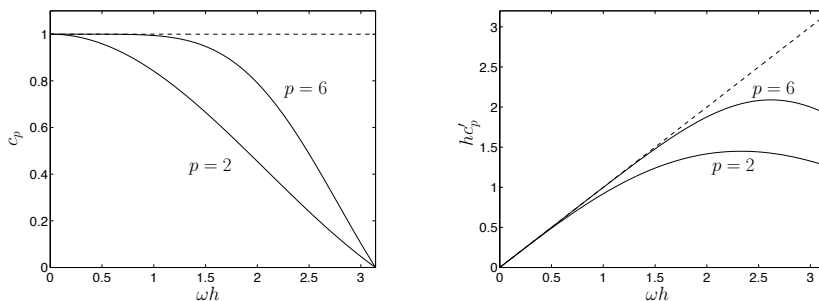


Figure 2.1. (left) The wave speed c_p for (2.1), as approximated by p th order central finite differences. The dashed reference line indicates the desired wave speed, which is 1 for all frequencies. (right) The wave speed c'_p for (2.2) scaled by the grid size, as approximated by p th order central finite differences. The dashed reference line indicates the desired wave speed ω .

We now contrast this with the Schrödinger equation for a free particle in one dimension,

$$u_t = i u_{xx}, \quad x \in \mathbb{R}, t > 0, \quad (2.2)$$

which has the plane wave solutions

$$u(x, t) = e^{i\omega(x - \omega t)}, \quad \omega \in \mathbb{R}.$$

The TDSE is a dispersive wave equation, i.e., the phase velocity of a wave depends on its spatial frequency. In the case of the TDSE, the phase velocity grows linearly with the frequency. As the number of wave lengths the solution is transported seems to be a measure of the difficulty of a wave propagation problem, this indicates that solving Schrödinger equations is harder than solving linear hyperbolic problems. Indeed, to keep the error below ε at time t , we need at least

$$M'_p = C'_p \left(\frac{\omega^2 t}{\varepsilon} \right)^{1/p}$$

points per wave length when solving the Schrödinger equation. This also indicates that the gain of using higher order methods is higher for Schrödinger problems. This last statement is supported by common practice: The methods used when solving the Schrödinger equation are most often of higher order than the methods used for hyperbolic problems. While methods of order four or six often are considered sufficient for hyperbolic problems, spectral and pseudospectral methods dominate the scene for the Schrödinger equation. The Fourier pseudospectral method [53, 55], which is particularly useful thanks to the fast Fourier transform (FFT), is equivalent to an (infinitely wide) infinite order finite difference method [19, 20].

2.2 Time integration

Linear hyperbolic problems are fairly well-behaved when it comes to time stepping. For short time simulation, many general-purpose explicit Runge–Kutta and linear multistep methods are adequate, given that a (usually mild) time step restriction is satisfied [14, 25]. For the Schrödinger equation, which is not hyperbolic since the speed of propagation is unbounded, standard time stepping schemes tend to perform poorly. We will discuss that in more detail later in this section.

It is common that the solutions of wave propagation problems have conserved quantities and symmetries. There may be much to win by using a time stepping scheme which takes such properties into account, especially for long time simulation. Such methods are studied in the field of geometric numerical integration [10, 32]. The canonical application area for geometric integration is Hamiltonian ODEs. A Hamiltonian ODE is time-reversible, and preserves a Hamiltonian functional, or energy, and a quantity known as its symplectic bilinear form. The workhorse of Hamiltonian ODEs, the Störmer–Verlet or leapfrog method [33, 91], is time-reversible and symplectic, i.e., it preserves the symplectic form. Symplectic methods do not preserve the Hamiltonian exactly, but almost. By backward error analysis of symplectic methods one can show that there exists a modified Hamiltonian, close to the true Hamiltonian, which the method preserves up to an error which is exponentially small in the time step [32]. In, for instance, molecular dynamics, such preservation properties are very important. For large systems and long simulation times it is often inevitable that the error may become very large in absolute terms, but if energy, momentum, and angular momentum are conserved to high accuracy the simulation may still retain many macroscopic properties of the system.

Both the wave equation and the Schrödinger equation are, with suitable boundary conditions, Hamiltonian PDEs. After appropriate discretisation in space the resulting systems of ODEs are also Hamiltonian. In addition, the Schrödinger equation preserves the L^2 -norm of the solution. As previously mentioned, standard explicit time stepping schemes such as the leapfrog meth-

od work well for the wave equation, given that a time step restriction $\Delta t \leq \alpha \Delta x$ is satisfied. The same holds in principle for the Schrödinger equation, but since it has a higher order derivative in space than in time, the time step restriction becomes $\Delta t \leq \alpha' \Delta x^2$. This is very restrictive, and renders standard time stepping schemes practically useless. Instead, different kinds of exponential integration methods dominate the scene. These include splitting methods [18, 64, 80, 89], Krylov subspace methods [4, 43, 56, 74] and polynomial (Chebyshev) methods [86]. Splitting and Krylov subspace methods are unitary, i.e., they preserve the L^2 -norm of the solution, but only the splitting methods are symplectic, and only symmetric splitting methods are time-reversible [59].

The simplest possible splitting method, the Lie–Trotter scheme [89], reads

$$e^{h(A+B)} \approx e^{hB} e^{hA},$$

and is first order accurate in the time step h . A and B are the operators occurring in the right-hand side of the differential equation; for (1.2) we get $A = (i/2m)\Delta$ and $B = -iV$. Higher order accurate splitting methods can be constructed by using more substeps in the decomposition [7, 32]. Splitting methods are particularly useful when Fourier methods are applied to the TDSE since the potential is diagonal in coordinate space, and the Laplace operator is diagonal in frequency space. Diagonal operators are trivially exponentiated, and the FFT provides a fast means for going between the coordinate and frequency representations. We can thereby compute the substeps in essentially linear time. As its substeps are unitary, any splitting scheme for the TDSE is unconditionally stable. Any consistent splitting method is exact if A and B commute, so naturally, commutators of A and B occur in the error estimates. Order conditions can be derived using the Baker–Campbell–Hausdorff (BCH) formula [32] or Taylor expansion, but the resulting error estimates are only valid for bounded operators A and B and sufficiently small time steps. By using more delicate techniques, one can derive error estimates which under certain assumptions are valid for PDEs and longer time steps. Error estimates for the Lie–Trotter and the second order Strang [80] schemes for the Schrödinger equation were derived in [46], in which bounds on the commutators was an important ingredient. Estimates for higher order splitting methods, valid for PDEs, were derived in [36, 67, 88]. In Paper IV we extend the error analysis from [88] to splitting schemes of force-gradient type [52].

3. High-dimensional problems

The dimensionality of the TDSE grows linearly with the number of particles under consideration. Solving high-dimensional PDEs numerically is notoriously difficult due to the exponential growth of work and data with the dimension when standard methods are applied. If n grid points are needed in one dimension, one generally has to expect that n^d grid points are required when a similar problem is to be solved in d dimensions. To be able to attack problems in more than, say, four dimensions, some kind of reduction of the model is necessary—solving the problem on a standard grid would be intractable.

3.1 Semiclassical methods and high frequency approximation

The TDSE (1.2) for molecular dynamics and Hamilton's equations (1.3) model the same physics: Classical dynamics can be seen as an approximation of quantum dynamics. A molecular system behaves more classically when the nuclei are heavy, and classical dynamics is retained in the $m \rightarrow \infty$ limit. By using this relation between quantum and classical dynamics, approximate methods can be constructed. Such methods are often called semiclassical. Many of these are based on Gaussian wave packets [38]. Gaussian wave packets stem from the observation that a complex Gaussian with appropriately evolved parameters solves the TDSE when V is a convex quadratic polynomial, the so-called harmonic oscillator, exactly. In particular, the centre point and centre frequency of the Gaussian are evolved according to (1.3), and its covariance matrix according to a Riccati equation. For anharmonic potentials, the TDSE is solved approximately by evolving the wave packet according to the second order Taylor expansion of the potential. For increasing m the wave packet becomes more narrow, and the error from the truncation of the Taylor expansion decreases. This justifies using Gaussian wave packets to construct a semiclassical method. The wave function also becomes more oscillatory as m increases. Gaussian wave packets with fixed width, so-called frozen Gaussians, are also popular [39, 40, 83]. One can construct approximations of higher order in m^{-1} by multiplying the Gaussian wave packet with a carefully constructed polynomial prefactor. These functions are known as Hagedorn wave packets [30, 31]. In [17], a practical computational algorithm using Hagedorn wave packets was devised. Such wave packets are an important ingredient in

Paper I, although not as a semiclassical method. A review of semiclassical methods, written from a mathematical perspective, is given in [47].

Gaussian wave packets have also been explored by the mathematical community under the name Gaussian beams [42, 58, 71, 72]. They were motivated as an asymptotic method in the limit of high spatial frequency, and can be derived from the eikonal and transport equations of geometrical optics. Application-wise, Gaussian beams initially focused on optic, acoustic and seismic wave propagation rather than on quantum mechanics, but they can be derived also for the TDSE in an analogous way. For the Schrödinger equation there is a close relation between high frequency approximation and semiclassics, as the wave function will have wave lengths of order $m^{-1/2}$. In Paper II, we couple Gaussian beams for the TDSE to a direct solver using finite differences on summation-by-parts form.

3.2 Sparse grids

A different way of reducing the computational complexity of solving high-dimensional PDEs is sparse grids. They were first introduced for multivariate quadrature and interpolation by Smolyak [79]. A multi-dimensional grid is constructed using a hierarchy of one-dimensional quadrature formulas of different spacing. Most of the n^d points occurring in the full grid are omitted, and the resulting grid has only $\mathcal{O}(n(\log n)^{d-1})$ points. Sparse grid techniques have also been used for partial differential equations [11, 96]. The accuracy of approximation on sparse grids relies on fast decay of mixed derivatives. This somewhat limits the applicability of the approach.

3.3 Low-rank approximation

Another way of reducing the model, which has had much success in quantum dynamics, is low-rank approximation. In two dimensions, where function values on a tensor product grid can be represented by a matrix, a rank- r approximation can be constructed using a truncated singular value decomposition,

$$\mathbb{R}^{n \times n} \ni A \approx Y = USV^T,$$

where $U, V \in \mathbb{R}^{n \times r}$ have orthonormal columns, and $S \in \mathbb{R}^{r \times r}$. When $r \ll n$, this significantly reduces the amount of data required to represent (an approximation of) the matrix. Various low-rank tensor formats generalise this data-sparse representation to higher dimensions [27, 51]. Of particular relevance is the Tucker format, which is used in the multi-configurational time-dependent Hartree (MCTDH) method [65, 66]. MCTDH and its variants have become standard approaches for quantum dynamics problems of moderately high dimension. The amount of data needed in the MCTDH method still scales exponentially with the dimension, but with a possibly much smaller base. For a

rank- r representation of a d -dimensional wave function with n points per dimension, $r^d + drn$ numbers must be stored. This exponential scaling is remedied in multilayer MCTDH [93, 94], which uses variants of the hierarchical Tucker (H-Tucker) format [28]. The r^d -element core tensor of the Tucker format is decomposed hierarchically, forming a tree structure. The H-Tucker format is retained as a special case when the tree is binary, and requires the storage of $\mathcal{O}(drn + dr^3)$ numbers. Another important special case is tensor trains (TT) [70], where the tree is binary and made as unbalanced as possible. The tree then gets a linear structure, which simplifies many algorithms. A tensor train requires the storage of $\mathcal{O}(dr^2n)$ numbers. Before being explored in the mathematics community tensor trains were discovered by physicists, who call them matrix product states [29, 75, 92].

The spaces of Tucker, H-Tucker and TT tensors of fixed rank are embedded manifolds in $\mathbb{C}^{n_1 \times \dots \times n_d}$ [45, 49, 90], which forms a foundation for stable computations. For low-rank approximation of time-dependent problems we apply the Dirac–Frenkel time-dependent variational principle [15, 54, 59]. Consider the tensor differential equation

$$\dot{A}(t) = F(A(t)), \quad A(0) = A_0 \in \mathbb{C}^{n_1 \times \dots \times n_d}.$$

If \mathcal{M} is the manifold of tensors of fixed rank and $T_Y \mathcal{M}$ its tangent space at $Y \in \mathcal{M}$, application of the Dirac–Frenkel principle gives a variational problem for the low-rank approximation $Y(t)$ of $A(t)$, reading, *for all $t \in [0, T]$, find $Y(t) \in \mathcal{M}$ such that $\dot{Y}(t) \in T_{Y(t)} \mathcal{M}$ and*

$$\langle X, \dot{Y}(t) \rangle = \langle X, F(Y(t)) \rangle \quad \forall X \in T_{Y(t)} \mathcal{M}, \quad Y(0) = Y_0 \in \mathcal{M}. \quad (3.1)$$

Quasi-optimality results for dynamical approximation with Tucker, H-Tucker and TT tensors are given in [3, 49, 62]. In [60, 61], an efficient time stepping scheme for (3.1) on the manifolds of low-rank matrices and TT tensors was presented. The scheme has some remarkable robustness properties, which we analyse in Paper VI.

4. Absorbing boundary conditions

The Schrödinger equation (1.2) is generally posed on the infinite spatial domain \mathbb{R}^d . This makes the numerical treatment of boundary conditions delicate in many cases. In other cases, it may become very easy. The easy cases are bound problems, where a localised wave packet oscillates around an equilibrium. The wave function then decays fast, often exponentially, away from a region of effective support, and we can safely truncate the domain. At the introduced boundary we can use any boundary condition we want, as long as it is well-posed and does not excite the system. A popular choice is periodic boundary conditions, as they are easy to implement, do not affect the stability of the discretisation, and facilitate the use of Fourier methods.

For scattering and dissociation problems, the situation is more complicated. There, the wave function will not be confined in a small region, but instead spread over an extended area. It is, however, reasonable also for such problems to truncate the domain outside the region where the phenomenon under study takes place. The scattered or dissociated wave packet spreading towards infinity is usually of lesser interest. In such cases it is important that the truncation of the domain does not affect the solution in the interior, the boundary should be absorbing so that a wave moving towards the boundary exits the domain without reflections. Exact absorbing boundary conditions can often be constructed in theory, but they are global in both space and time and therefore of limited use for practical computations. Local absorbing boundary conditions were pioneered by Engquist and Majda for the scalar wave equation [16]. They related spatial and temporal derivatives of the solution at the boundary to derive local absorbing boundary conditions of different orders, which yield well-posed initial-boundary value problems. The boundary conditions absorb waves with normal incidence exactly, but give reflections when waves approach tangential incidence. The higher order boundary conditions deteriorate slower when the angle of incidence grows, but involve higher order derivatives and are therefore more cumbersome to implement. Engquist–Majda-type absorbing boundary conditions have also been constructed for the Schrödinger equation, see, e.g., the review article [1].

An alternative to local absorbing boundary conditions is to embed the domain in an absorbing layer where a modified equation is solved. One way of modifying the equation is to add an imaginary term to the potential, a so-called complex absorbing potential [63, 68, 73]. This technique is popular among chemists. Another technique, which we employ in Papers II and III, is perfectly matched layers (PML). It was first invented for Maxwell's equations

in [6], and has been used and studied a lot for hyperbolic problems [2, 5]. The interface between the regions where the original PDE and the PML equations are to be solved does not cause any reflections—the absorbing layer is perfectly matched. If the layer is infinitely wide we have a perfectly absorbing boundary at the interface. That is, of course, of little practical use as the computational domain then still is infinite, but since waves are damped within the layer we only get small reflections when the layer is truncated to finite size. Some small reflections may also arise due to the discretisation of the PML equations. In this thesis, we use the PML developed for the Schrödinger equation in [69]. An attractive feature of this PML is that it only amounts to a modification of the kinetic energy operator in the layer. For hyperbolic problems, auxiliary variables with accompanying equations have to be introduced in the PML.

5. Summary of papers

5.1 Paper I

In this paper we develop an adaptive pseudospectral method using Hagedorn wave packets [30, 31]. Hagedorn wave packets are Gaussian functions multiplied by polynomial prefactors. They are orthonormal in $L^2(\mathbb{R}^d)$ and generalise Hermite functions to multiple dimensions. Similarly to Gaussian wave packets, their position and shape parameters can be evolved in time such that each basis function solves the harmonic oscillator exactly. A difference compared to Gaussian wave packets is that all the Hagedorn basis functions have the same position and shape parameters, while the Gaussian basis functions move independently of each other. Instead, the Hagedorn basis functions differ by being of different polynomial degree. When evolved on an anharmonic potential, error estimates in terms of m^{-1} can be derived. A practical computational algorithm, designed for the semiclassical regime, was developed in [17].

Error estimates for semiclassical methods deteriorate with time, in some cases rather quickly. In this work we use Hagedorn wave packets in a non-semiclassical setting. Instead of letting the basis follow the classical equations of motion, we make it match the support of the wave packet. This is done using a proportional-derivative controller, a standard tool from automatic control. The end result is a collocation method with a time-dependent, adaptive basis. Semiclassical error estimates for Hagedorn wave packets are no longer valid, but instead we get the possibility for accurate simulation over longer periods of time in situations where we can afford to resolve the wave function. We demonstrate applicability of the method on photodissociation problems in one and two dimensions.

5.2 Paper II

In the semiclassical regime, i.e., when the particles are heavy, solutions to the TDSE are highly oscillatory. This makes grid-based methods very expensive also in low dimensions, and it might be worthwhile to consider alternative approaches, e.g., asymptotic methods. One such asymptotic method is Gaussian beams, cf. Section 3.1. Gaussian beams are accurate in the high frequency regime, where the wave length is much shorter than the scale on which the potential has variations. In this paper we consider a problem where a local sharp

feature of the potential breaks the validity of high frequency asymptotics. Such a feature could be, e.g., a reaction barrier allowing for tunnelling, which is a feature Gaussian beams cannot represent. We develop a hybrid method which uses Gaussian beams in smooth regions and a finite difference method where the potential has sharp features. A similar hybrid method was developed in [48]. Our method is based on a similar idea, but is more efficient since it uses thinner interfaces which reduces redundancy, and also uses cheaper algorithms for handling the interfaces.

The method is based on an *a priori* decomposition of the domain into regions handled by Gaussian beams and finite differences, respectively. This is justified since the choice of method depends on what the potential looks like, which is information available before starting the computation. The main contribution of this paper is the treatment of the interfaces. We treat the transformation of Gaussian beams to a grid representation using Huygens' surfaces [85, Ch. 6.5], which is a standard approach in electromagnetics but little used for other types of problems. Transforming the grid representation to Gaussian beams is less straight-forward. In [87] it was shown how a highly oscillatory function in WKB form, i.e., of the form

$$u(x) = A(x)e^{i\phi(x)/\varepsilon}$$

with A and ϕ independent of $\varepsilon \ll 1$, could be written as an integral superposition of Gaussian beams, and error estimates in terms of ε were proven. We treat the grid-to-beams transformation in a similar way, but using a superposition over the temporal instead of the spatial coordinate. We also prove a similar error estimate. When the outgoing wave packet has crossed the point where it is translated to Gaussian beam representation, we remove it from the grid by absorbing it with a PML.

5.3 Paper III

This work was motivated by a particular application in experimental physics, known as higher harmonic generation (HHG) [95]. In a variety of experiments, sequences of short but highly energetic pulses of light are desired. HHG is a means of creating such pulses. This is done by exposing a thin atomic gas to an oscillating electric field created by a strong laser beam. The electric field may then, with some probability, rip one valence electron off, ionising the atom. Since the field oscillates, the electron may be accelerated back to recombine with the ion. As it returns to a bound orbit, the excess energy it has acquired from the field will be emitted as a high energy photon. Since the electric field oscillates with a certain frequency, the atomic gas will emit bursts of light at regular intervals.

A difficulty with solving electron dynamics problems numerically is the lack of regularity of the solution, which is caused by the singular Coulomb

potential. By formulating the problem in cylindrical coordinates, using Bessel functions in the radial and a Fourier basis in the axial direction, the situation is somewhat improved, but the potential still has a discontinuous first derivative. This results in a jump in the third derivative of the solution in the axial direction. We derive an expression for the size of this jump, and propose a regularisation which cancels it. One would expect this to increase the order of accuracy from two to three, but numerical experiments indicate that the method is fourth order accurate. To explain this behaviour we study the Laplace transform of the error equation. By exploiting localisation of the error, both in time and frequency, we are able to prove a sharp error estimate.

5.4 Paper IV

This paper concerns splitting methods for, primarily, Schrödinger type problems. We consider a problem of the form

$$u_t(t) = Au(t) + Bu(t), \quad u(0) = u_0,$$

posed on a Banach or Hilbert space, with possibly unbounded operators A and B . If additional assumptions are made, this can sometimes be exploited to construct more efficient methods. For example, if the iterated commutator $[B, [B, [A, B]]]$ vanishes, which is the case for the TDSE, splitting schemes of Runge–Kutta–Nyström type [7, 32] can be applied. By exploiting that this commutator vanishes they need fewer stages for the same order of accuracy, compared to standard splitting methods. In this paper we assume this, and also that an explicit, computable expression for the commutator $[B, [A, B]]$ is available. One can then use force-gradient, or modified potential, splitting methods [52, 81, 82]. For the Schrödinger equation, with $A = i\Delta$ and $B = -iV(x)$, we get $[B, [A, B]] = 2i|\nabla V|^2$, i.e., the commutator has the form of a potential.

Classical error analysis for splitting methods can be based on the BCH formula [32] or Taylor expansion. That is, however, only valid if A and B are bounded operators and the time step h is sufficiently small. Error estimates which hold also for unbounded operators, such as differential operators, are called stiff error estimates. Stiff convergence for standard splitting methods of any order, applied to the Schrödinger equation, was proven in [88]. In this paper we extend that result to force-gradient methods.

5.5 Paper V

For wave propagation problems, high order methods are advantageous. For p th order finite difference or finite element methods, the cost of numerical

differentiation is linear in the number of grid points, with a p -dependent proportionality constant. The error decays as Δx^p when the spatial stepsize Δx is decreased. For spectral and pseudospectral methods on the other hand, the error decays faster than any polynomial in Δx , while the computational cost generally is quadratic in the number of grid points. This quadratic scaling easily renders spectral methods prohibitively expensive. There are, however, situations where the computational cost of numerical differentiation can be drastically mitigated. The most prominent example is periodic problems, where the Fourier method can be applied. With the help of the fast Fourier transform, the cost is then again essentially linear. For problems with less trivial boundary conditions it is not as easy to make spectral methods efficient.

In this paper, we develop a spectral method which is applicable to time-dependent PDEs with a wide range of boundary conditions, and which scales essentially linearly with the number of grid points. The method is based on a Galerkin ansatz in a basis of Legendre polynomials and operates in the spectral space, i.e., we work with the expansion coefficients for the Legendre polynomials rather than with the point-wise values of the solution. The motivation for this is that spatial derivatives then can be evaluated in linear time using recurrence relations, see, e.g., [12, Ch. 2.3]. This however comes at a price—multiplicative operators such as variable coefficients become global, and the cost for applying them will be quadratic in the number of grid points. In [17] it was suggested how this could be circumvented when the coefficients are much smoother than the solution, in the context of solving the Schrödinger equation in a basis of Hermite functions. The method was further elaborated on, and analysed in detail, in [9]. It uses orthonormality and recurrence relations for the basis functions to construct a recursive procedure for how to apply polynomial multiplicative operators. If the variable coefficients are much smoother than the solution they can be approximated by polynomials of relatively small degree, which is the key to the efficiency of the approach. We extend this approach to initial-boundary value problems, where we impose boundary conditions weakly using a penalty technique [13, 21, 22, 41]. The penalty terms are global in the spectral space, but they are also of low rank which makes it possible to evaluate them in linear time. We prove that the method is stable and spectrally accurate for the scalar wave equation.

5.6 Paper VI

A difficulty with dynamical low-rank approximation is that the curvature of the approximation manifold \mathcal{M} grows without bound when singular values taken into account in the approximation tend to zero. The dynamical low-rank approximation, as given by the Dirac–Frenkel principle (3.1), can be written equivalently as

$$\dot{Y}(t) = P(Y(t))F(Y(t)), \quad Y(0) = Y_0 \in \mathcal{M}, \quad (5.1)$$

where $P(Y)$ is the orthogonal projection onto the tangent space $T_Y\mathcal{M}$. The Lipschitz constant of $P(Y)$ is inversely proportional to the smallest nonzero singular value of Y . This forces standard time stepping schemes to take very small time steps. Small singular values in Y is something one has to expect. In the initial data Y_0 we have neglected all singular values which are smaller than some threshold. However, the singular values of the matrix or tensor¹ we want to approximate typically decay without a distinct gap. If the largest neglected singular value is not small we will neglect a lot of information, and thus make a significant modelling error. There is, however, no reason to expect the smallest included singular value to be much larger, and as we just saw, small included singular values causes problems with large Lipschitz constants. Small singular values in the low-rank approximation is therefore most often something we desire, but at the same time something we would rather avoid.

Recently, an efficient time stepping scheme for low-rank approximation of matrix and TT-tensor differential equations was proposed [60, 61]. This scheme is a splitting method, based on decomposing the projection $P(Y)$ into $2d - 1$ terms. Each substep of the projector-splitting integrator is an evolution equation for at most r^2n degrees of freedom. Furthermore, if $F(Y)$ is linear the subproblems will also be linear, even though (5.1) is nonlinear. Standard error estimates for splitting methods [32] will break down when singular values tend to zero, as they depend on the Lipschitz constant of P . This deterioration in accuracy is however not seen in numerical experiments. It was proven in [60, 61] that if $F = F(t)$ does not depend explicitly on A , and if $A(t)$ is of the prescribed rank, the projector-splitting integrator is exact. This is an indication of the remarkable robustness of the method.

In this paper, we present a thorough robustness and accuracy analysis of the projector-splitting integrator. We prove that if the exact solution is almost of the prescribed rank, i.e., if F maps onto the tangent bundle of \mathcal{M} up to an ε -perturbation, the error $\|A(t_k) - Y(kh)\|$ will be bounded in terms of ε and h , where h is the time step. The error estimate depends on the Lipschitz constant of F , but not on the Lipschitz constant of P or in any other way on the included singular values. We prove the result by combining the exactness result from [60, 61] with the fact that the spaces spanned by the basis vectors for each coordinate direction are preserved in all but one substep of the splitting scheme. Using that, we can avoid the large Lipschitz constants of the projections when we isolate the $\mathcal{O}(\varepsilon)$ perturbations.

¹The singular values of a tensor should in this context be understood as the singular values of its matrix unfoldings [23, 70].

6. Sammanfattning på svenska

Vår uppfattning av världen omkring oss formas i hög utsträckning av vågutbredningens fysik, då vi med synen och hörseln uppfattar optiska respektive akustiska vågor. Oftast får vi därigenom en någorlunda rättvisande beskrivning av verkligheten, men ibland kan egenheter hos vågfysiken lura oss. Exempelvis gör dopplereffekten att samma fordon låter annorlunda när det närmar respektive avlägsnar sig, och regnbågen är en effekt av att ljushastigheten i vatten beror på ljusets våglängd. Vågor används i ett otal tekniska tillämpningar. Etermedia och mobiltelefoni sänds med elektromagnetiska vågor, dvs. vågor av samma sort som synligt ljus, men med mycket större våglängd. Ett ekolod använder vanliga ljudvågor för att mäta havets djup, på samma sätt som fladdermöss använder ekot från sina rop för att hitta flygande insekter. Den här avhandlingen handlar om beräkningsmetoder för vågutbredning.

Många vågutbredningsproblem är i grunden ganska enkla. Ljudvågor är små svängningar i lufttrycket som förflyttar sig med ljudhastigheten c . Den matematiska beskrivningen av ljudutbredning, vågekvationen, är också enkel. Den lyder

$$\begin{aligned}u_{tt} &= c^2 \Delta u, & x \in \Omega, \\ n \cdot \nabla u &= 0, & x \in \partial\Omega,\end{aligned}\tag{6.1}$$

där $u = u(x, t)$ är lufttryckets avvikelse från jämviktsläget vid positionen x och tiden t , och Δ är Laplaceoperatoren. Randvillkoret $n \cdot \nabla u = 0$ beskriver reflektion mot hårda väggar. I en fri luftmassa är det betraktade området $\Omega = \mathbb{R}^3$, och problemet låter sig enkelt lösas med variabelseparation. Om Ω beskriver en mer komplicerad geometri, exempelvis en konsertsal, blir lösningen mycket mer komplicerad eftersom ljudvågorna reflekteras mot rummets golv, väggar och tak. Problemet kan lyckligtvis fortfarande lösas numeriskt, med hjälp av datorberäkningar.

Ett centralt koncept inom kvantmekaniken, våg-partikel-dualiteten, innebär att vi inte bara uppfattar världen omkring oss genom vågor: världen består av vågor, en slags sannolikhetsvågor. Det är omöjligt att veta exakt var en partikel befinner sig, vi kan bara känna till en sannolikhetsfördelning för partikelns position. Sannolikhetsfördelningen kan beräknas med hjälp av den så kallade vågfunktionen. Vågfunktionen uppfyller Schrödingerekvationen [78],

$$iu_t = -\Delta u + V(x)u, \quad x \in \mathbb{R}^n, t > 0,\tag{6.2}$$

som är den ekvation vi oftast studerar i den här avhandlingen. Schrödingerekvationen är av andra ordningen i rummet och första ordningen i tiden; den

imaginära enheten gör att lösningen trots detta har vågkaraktär. Skillnaden i ordning på tids- och rumsderivatorna gör att vågor med kortare våglängd transporteras snabbare, dvs. Schrödingerekvationen är en dispersiv vågekvation. Detta gör det besvärligare att lösa (6.2) med en dator, jämfört med (6.1).

Det är vanligt att vågor rör sig över långa sträckor, ofta tiotals eller hundratals våglängder. Ett vanligt problem vid numerisk lösning av vågutbredningsproblem är dispersionsfel, att man får ett litet fel i utbredningshastigheten. Några hundra våglängder bort kan ett fel på en halv våglängd tyckas bagatelartat, men om vågen där, t.ex., samverkar med en annan våg kan det få stora konsekvenser om vågen pekar upp istället för ned vid en viss punkt. Schrödingerekvationen är extra känslig för den här typen av fel eftersom den transporterar vågor med kortare våglängd snabbare. Högre ordningens metoder, som har små dispersionsfel, används ofta för vågutbredningsproblem. Spektralmetoder, i synnerhet Fouriermetoden, är särskilt populära för Schrödingerekvationen. Vi använder spektralmetoder i flertalet delarbeten. I Manuskript III utvecklar och analyserar vi en vidareutveckling av Fouriermetoden för problem med stygga lösningar, och i Manuskript V utvecklar vi en spektralmetod för problem med icke-triviala randvillkor.

Att (6.2) har en högre ordningens derivata i rummet än i tiden gör också att den blir styv. Det innebär att man måste ta väldigt små steg om man propagerar lösningen i tiden med vanliga metoder för tidsstegning, t.ex. explicita Runge–Kutta- eller flerstegsmetoder [34]. Sådana metoder är väldigt ineffektiva för Schrödingerekvationen. Istället används ofta olika former av exponentialintegratorer, t.ex. splittingmetoder [18, 64, 80, 89], Krylovmetoder [4, 43, 56, 74] och Chebyshevmetoder [86]. I Manuskript IV bevisar vi en rigorös feluppskattning för en klass av splittingmetoder.

Ytterligare en svårighet med Schrödingerekvationen är dess höga dimensionalitet. För traditionella, nätbaserade, beräkningsmetoder växer arbete och minnesåtgång exponentiellt med dimensionaliteten. Ett system med N partiklar beskrivs av en Schrödingerekvation i $3N$ rumsdimensioner. Om vi använder n (ofta något hundratal) nätpunkter per dimension får vi totalt n^{3N} punkter i nätet. Detta blir snabbt ohållbart. Genom att bara betrakta partiklarnas rörelser i förhållande till varandra kan vi få ned dimensionaliteten en aning, $3N$ reduceras till $3N - 6$, men lösning på nät är fortfarande begränsat till mycket små system.

I Manuskript VI betraktar vi ett sätt att möjliggöra simulering av större system: lågrangsapproximationer. Genom att utnyttja att lösningen (förhoppningsvis) har en särskild struktur kan vi reducera mängden arbete avsevärt. Istället för att lösa ett d -dimensionellt problem löser vi r^2d endimensionella problem, där $r \ll n$ är lösningens rang, och kopplar samman de endimensionella lösningarna på ett finurligt sätt. Mängden data reduceras därmed från n^d till r^2dn , vilket är en himmelsvid skillnad. I [60, 61] presenteras en tidsstegningsmetod för lågrangsapproximationer som är förvånansvärt robust. Traditionell felanalys bryter samman när singularvärden i lösningen närmar sig noll,

men den här metoden ger bra resultat även i sådana fall. I Manuskript VI tar vi ett stort steg på vägen till att förstå varför. Vi bevisar en feluppskattning som är oberoende av lösningens singularvärden.

Author's contributions

Paper I

The author of this thesis had the main responsibility for preparing the manuscript and performed all computations. The ideas were developed by the author in consultation with the co-authors.

Paper II

The author of this thesis had the main responsibility for preparing the manuscript and performed all computations. The ideas were developed in close collaboration between the authors.

Paper III

The author of this thesis is the sole author of the manuscript.

Paper IV

The author of this thesis is the sole author of the manuscript.

Paper V

The work was done in close collaboration between the authors. The author of this thesis made significant contributions in particular to the treatment of boundaries and to the error analysis.

Paper VI

The author of this thesis contributed substantially to the proof for the matrix case, most importantly with the idea of combining the exactness result with the preservation of singular vectors. He also contributed with large parts of the text and conducted several of the numerical experiments.

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