Chapter 1

Introduction

In this thesis we present theoretical studies of:

1. Optimal control of a quantum system with relaxation over a finite time interval,
2. Excitation spectrum of interacting electrons confined in a quantum dot,
3. Optimal control of the carrier dynamics in nanostructures using a double quantum dot as a model device,
4. Strong excitation and consequent explosion of noble gas clusters interacting with intense femtosecond laser pulses.

To understand the motivation for these problems, which all have in common the dynamical behavior of nanoscale systems at a non-equilibrium, we describe first recent advances and studies of the underlying physics in more details.

In the last decade the development of laser systems opened the way for creation of ultrashort femtosecond pulses with controlled shape, spectrum and polarization [1]. This can be used as an ideal tool to manipulate quantum objects. For instance, with the help of the optimal control field, one can induce chemical reactions which are not possible or very difficult to carry out [2].

After the seminal work of Judson and Rabitz [2], where the authors suggested a variational formulation of optimal control in quantum systems and a procedure to solve this problem, many theoretical and experimental investigations were devoted to the problem how "to teach" lasers to drive molecular reactions in real time [3, 4, 5, 6]. The idea consists in using pulse-shaping techniques to design pulses or sequences of pulses having a given optimal shape (and phase) so that the desired nuclear wave packet dynamics is induced. Optimal control of the internal motions of a molecule is achieved by exploiting a variety of interference effects associated with the quantum mechanics of molecular motion. Thus, the population of a certain vibrational state, which may be responsible for the yield of a chemical reaction, can be controlled.

Using a variational formulation of the control problem it was shown, how to construct optimal external fields (e.g. laser pulses) to drive a certain physical quantity, like the population of a given state, to reach a desired value at a given time [7, 8, 9]. However, even for the simplest control problems the obtained fields usually have a rather complex nature and cannot be easily interpreted [10]. Furthermore, since the optimal field arises as a solution of a system of coupled nonlinear integro-differential equations, which are treated numerically by application of iterative methods, there is also a problem related to the multiplicity of the obtained optimal solutions which are local extrema of the control problem [11]. Therefore, it is necessary to develop a new theory, which permits to derive analytical solutions for the optimal fields, which would guarantee their uniqueness.

Although the maximization of a given objective at a certain moment is relevant for many purposes, a more detailed manipulation of real systems may require the control of physical quantities during a finite time interval. An interesting example of such optimal control was recently performed on a system of shallow donors in semiconductors [12]. Using pulses of various shapes and duration one can control the photo-current in the system, while the total transferred charge is proportional to a time integral over the occupation of a certain excited state. The search for optimal fields able to perform such control is a highly complex problem for which no theoretical description has been given so far.

In many situations the controlled system cannot be treated as isolated, therefore dissipative and relaxation processes (due to the coupling to the environment or due to the contact with measuring devices) could play a significant role. In this case some limits of the optimal control of the system should exist [13]. The question is to estimate these limits quantitatively. An interesting problem usually not mentioned, is: "If the optimal control field for a quantum system without relaxation is obtained, how should it be modified in a presence of relaxation in the system?" In the presented work we develop a new formulation of the optimal control problem in quantum systems. Using an analytical approach we derive a differential equation for the optimal control field which we solve analytically for some limiting cases. The approach permits us to investigate optimal control of simple quantum systems with relaxation.

Another interesting problem in femtosecond physics is the interaction of intense femtosecond laser pulses having peak intensities up to $10^{15} - 10^{21}$ W/cm^2 with atomic clusters. For clusters containing tens, hundreds or thousands of atoms very different physical and chemical properties with respect to a bulk material appear and this depends crucially on their size. The interaction of intense femtosecond laser pulses with atomic clusters gives rise to a variety of fascinating phenomena not previously observed in similar experiments on atoms, small molecules or solids. For instance, the emission of x-rays and "hollow" atoms [14], hot electrons and ions [15, 16,

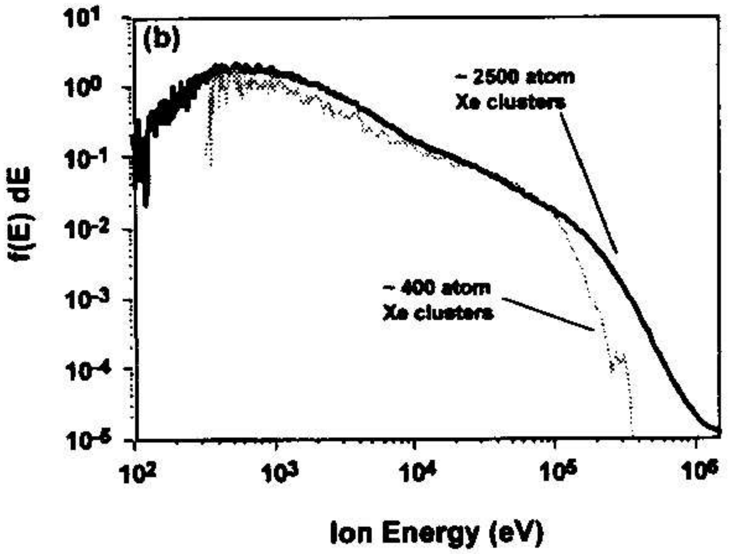

Figure 1.1: Measured distribution of the ejected ions as a function of their kinetic energy. Note the high energy tail appears for the cluster size ≈ 2500 atoms.

17], highly charged ions [16, 17, 18, 19, 20, 21, 22, 23] and monochromatic neutrons [24] were recently reported. Interaction of atomic clusters with intensive laser pulses even makes possible nuclear fusion using table-size installation [24, 25]. All these effects are related to the explosion of the clusters due to the absorption of an enormous amount of energy. Despite very intensive experimental and theoretical research [26, 28, 29, 30, 31, 32, 33, 34] on cluster explosion important questions still remain open. For example, the explosion mechanism of noble gas clusters is still a subject of controversy.

Ditmire and coworkers [16, 18] measured the kinetic energy distribution of Xe^{q+} ions produced upon explosion of Xe_N clusters (N varies from 10^2 to 10^5) and found that for clusters having initially a diameter larger than 65 $\AA$ ($N \approx 2500$) these distributions showed tails with highly energetic ions with energies up to 1 MeV (see Fig [1.1]). The measured charge state of such ions is up to +40. This result is remarkable, because it means that less than 30 % of electrons remain in the atom after the excitation. The ejected electrons exhibit kinetic energies up to 3 keV. To explain these facts a so called microplasma model was suggested [16], in which electrons in the cluster were treated as a classical plasma. It was assumed that during the explosion cold heavy ions are accelerated by the pressure of expanding hot electrons. This model predicts a linear dependence of the kinetic energy of the ejected ions on their charge state. Results of measurements [18] for large clusters were interpreted as a clear indication

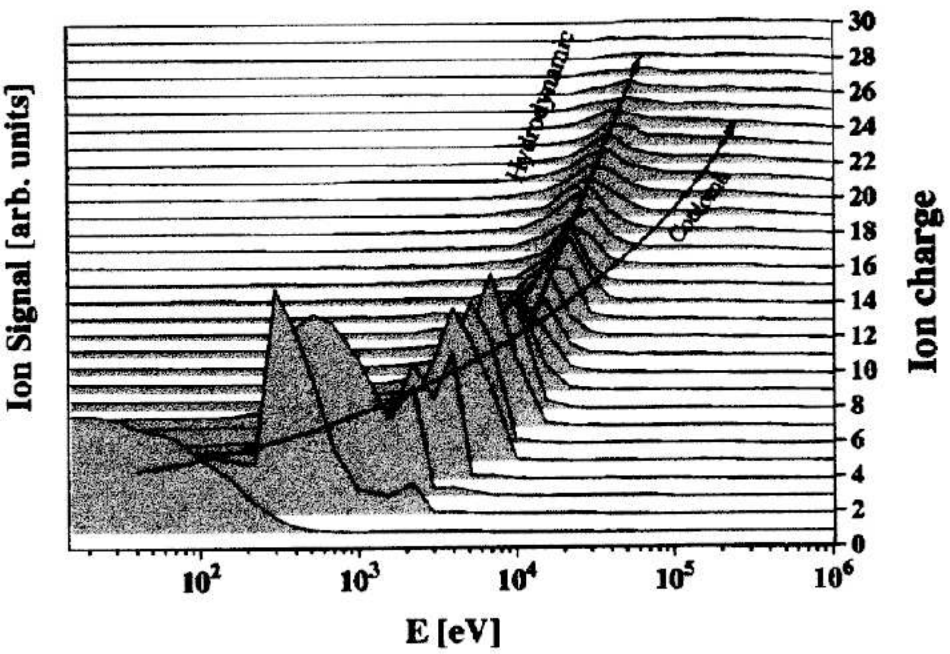

Figure 1.2: Measured dependence of the kinetic energy and ionization of the atoms after cluster explosion [17]. Note a strong correlation between energy of the ions and their charge state.

that the explosion is of hydrodynamic character.

On the other hand, Lezius and coworkers [17] measured the distribution of emitted Xe^{q+} ions as a function of their kinetic energy E_{kin} and charge q (see Fig. [1.2]). These experiments were performed on cluster ensembles containing mainly very large Xe_N for which the hydrodynamic mechanism should clearly dominate. However, the authors [17] claimed that by connecting the peaks of the distributions they obtained a superposition of two curves, fitted by the dependences $E_{kin} \propto q$ (hydrodynamic mechanism) and $E_{kin} \propto q^2$ (Coulomb mechanism)(see Fig. [1.2]). Interestingly, the latter one described the highly energetic ions in a clear contradiction with respect to the conclusions of Refs. [16] and [18]. Moreover, it seems, that the authors [17] did not mention that for distributions plotted in Fig. [1.2] they used the logarithmic scale for the energy E_{kin}. And the correct conclusion should be made: $\log(E_{kin}) \propto q$ and $\log(E_{kin}) \propto q^2$. Therefore, we conclude, that the cluster explosion mechanism is not understood well. In order to understand the mechanism of cluster explosion we suggest a new microscopic theory based on density-functional approach. This approach permits us to quantify the contribution of Coulomb and hydrodynamic mechanisms and explains the experimental data.

Within the same time period, during which the physics of the femtosecond laser fields exhibited its great success (80th-90th), mesoscopic physics advanced as a new era in physics and technology. Fabrication of mesoscopic systems and nanostructures (like clusters, nanowires, quantum dots, etc.) and further investigations of their properties represents technological breakthrough in many areas. For example, a further development of quantum dots (QD), which are often described as artificial solid-state atoms, provides a possibility to develop a new generation of semiconductor lasers. In these studies the semiconductor dots and rings are made from indium arsenide embedded in gallium arsenide [35]. They were grown using techniques developed within the past decade that allows much smaller nanostructures to be created. Such studies provide new perspectives on the internal quantum-mechanical workings of quantum dots. The ultimate goal is to create useful electronic and optical nanomaterials that have been quantum-mechanically engineered by tailoring the shape, size, composition and position of various quantum dots.

Excited electronic states in quantum dots persist for a relatively long time because they interact in a very restricted way with their environment. Normally, such interactions lead to decoherence or destruction of the quantum state. As a result, quantum dots may provide an excellent solid-state system for exploring advanced technologies is based on quantum coherence. For example, it may be possible to create and control superimposed or even entangled quantum states using highly coherent laser stimulation [57]. External control of the full quantum wavefunction in a semiconductor nanostructure may even lead to revolutionary new applications, such as those involving quantum computing, making computation in orders of magnitude faster than it is possible today.

The most complete and clear information about fabricated QD is contained in its excitation spectrum. Therefore the primary interest of experimentalists and theoreticians is about ground and excited states of the artificial atoms. For example, in the last years many measurements were performed on the conductivity of quantum dots. Connecting QD to current and voltage contacts allows the discrete energy spectra to be probed by charge-transport measurements [36]. Another way to obtain information about spectra of quantum dots based on optical spectroscopy measurements [37].

In the last years an increasing number of theoretical studies of the excitation spectra of quantum dots has been performed. One should mention studies using exact diagonalization [38], the variational method [39], diffusive Monte Carlo and the imaginary-time propagation [40], Density Functional Theory calculations [41, 42] and Path Integral Quantum Monte Carlo simulations [43]. Some special analytical solutions for two and three interacting electrons confined in two dimensions by harmonic potential were found [44].

All above mentioned methods have serious drawbacks because of computational difficulties which are increasing rapidly with the number of particles in the system, except, may be, Path Integral Quantum Monte Carlo calculations. However, this method has also a fundamental problem, the so called “sign problem” for fermionic systems, that leads to a very slow convergence of the method. Therefore, one can conclude, alternative methods to deal with quantum many body problems are desired. Note, that besides various applications to nanostructures, the search for the spectrum of a system containing strongly interacting particles represents by itself a fundamental problem in quantum theory. As an alternative approach to solve this problem we develop in this work a new method based on Genetic Algorithms, which have been successfully applied to solve multidimensional complex problems [5, 45, 46, 47, 48]. We test our method on the solution of various quantum few body eigenstate problems and find a very good convergence in all considered cases.

Because the underlying principles of quantum interference control of molecular dynamics are broadly applicable, it is a very attractive idea to combine femtosecond dynamics and mesoscopic physics and to perform coherent control on mesoscopic objects. For such systems, not the nuclear, but the electronic degrees of freedom might offer the possibility of control by pulse shaping. An important requirement for optimal control of quantum dynamics is the existence of phase coherence over a time range comparable to the duration of the pulse sequence. This condition can be fulfilled by mesoscopic systems like quantum dots. Many of the quantum-dot systems currently being studied have the potential to be combined into molecular complexes with one-, two- or even three-dimensional structures. One can imagine growing these solid-state atoms or molecules within structures containing electronic or magnetic gates and optical cavities, perhaps all connected together by quantum wires. One of the primary interest of scientists is the investigation of the currents in such systems resulting due to interaction with external coherent fields.

Coherent control of carrier dynamics in mesoscopic systems using external ultrashort time-dependent fields has become a subject of active research in recent years [12, 49, 50, 51]. In particular, the study of photon induced and photon suppressed quantum dynamical tunneling [52] has attracted much attention due to potential applications of these effects in quantum computing. Sequences of control pulses of different durations affect the systems in different ways and permit to perform some restricted manipulation of physical quantities, like the photon-induced current [12, 50]. In this work we study the possibility of the optimal control in nanostructures. Using a double quantum dot system as a model device we determine the optimal control fields which maximize the transferred charge in the system. The optimal solutions were obtained using Genetic Algorithms. We also investigate the influence of relaxation and decoherence in the system on the shape of the optimal fields.

In summary, despite the great success in mesoscopic physics there are still challenging theoretical problems not solved yet. Many questions regarding equilibrium and non-equilibrium dynamics of nanostructures are still open. The microscopical description of nanostructures involves quantum many body phenomena. This leads to difficult theoretical and computational problems which are not accessible within usual methods. Therefore, novel methods for these problems must be developed. In the considerable part of this work we use a genetic algorithm which is a very powerful and flexible numerical technique. This study should not only predict and explain various features of the systems, but should also give a possibility to control their dynamics. Solving the problems studied in this thesis, one can learn about transient states and time evolution of the complex few body quantum systems.

The present study consists of four parts:

1. In the first section an analytical theory for optimal control over a finite time interval in quantum systems with relaxation is presented. In this theory control of the objective at a given time is considered as a special case. By using a variational approach high-order Euler-Lagrange equations were derived, from which the optimal control fields are obtained analytically for some limiting cases. This approach guarantees the uniqueness of the obtained solution. Using the adiabatic approximation for the control field, an approximate analytical solution for the density matrix is derived. The theory is applied to two level quantum systems.

2. In the second section a new theoretical method based on evolutionary (genetic) algorithms is developed. This permits to determine efficiently the ground and exited states of few electrons confined in a quantum dot. This approach uses also the variational principle. We use a variational formulation for the ground state energy and for the partition function. To perform the search for the fittest wave function, the genetic algorithm (GA) is extended to treat quantum problems. It will be shown that this approach yields a very good convergence in all considered cases.

3. The third section of the theory is devoted to the optimal control of the carrier dynamics in nanostructures. The electron tunneling induced by an ultrashort electric field between two metallic contacts and coupled through a double quantum dot is analyzed theoretically. The equations of motion for the reduced density matrix to determine the transferred charge, which is a functional of the external field are solved numerically. Then, using the genetic algorithm the optimal shape of the electric field which maximizes the transferred charge is determined. The cases of zero and infinite interdot Coulomb repulsion are analyzed.

4. In the fourth section the explosion of Xe clusters due to the interaction with intense laser pulses is investigated. Using a microscopic theoretical approach we analyze the ultrafast ionization and explosion of

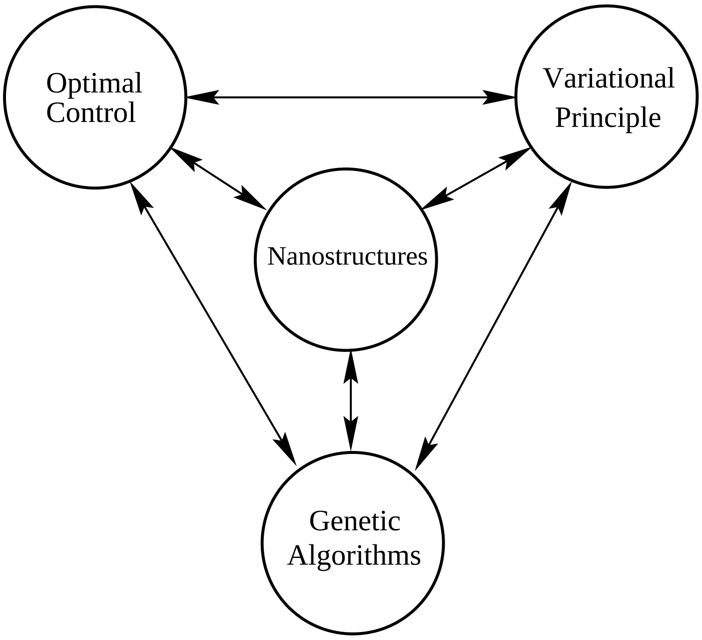

Figure 1.3: Relationship between the presented studies. In the first part we study the **optimal control** problem using Lagrangian (**variational**) formulation. In the second part we search for the excitation spectra of quantum systems using **variational principle** for the partition function and using **Genetic algorithms** to solve the problem. Possible applications to semiconductor **nanostructures** (quantum dots) are considered. In the third part we solve the **optimal control** problem in **nanostructures** using **Genetic algorithms** in order to find the optimal shape of the control field. And in the fourth part the ultrafast dynamics of atomic **nanostructures** (clusters) under strong femtosecond fields is investigated. The initial state of these clusters was determined using **Genetic algorithms**.

clusters induced by intense femtosecond laser pulses. It is shown, that the remarkable correlation between kinetic energy and charge of the produced ions observed in different experiments can be described as arising from strongly inhomogeneous charge densities in the clusters, which are induced by the strong quantum dynamics of the electrons after the laser excitation.

The internal relationship between these studies is illustrated in Fig. [1.3].

In Chapter 2, a detailed description of the used theoretical methods are given. Chapter 3 presents results of the analytical and numerical calculations. Finally, in Chapter 4, we discuss results, possibilities for further study and application of the developed theory.