Chapter 4

Conclusions and outlook

In this work we have developed and presented various theoretical models and methods in order to describe the following physical problems: optimal control of quantum systems for a finite time interval, calculation of the excitation spectrum of few-body quantum systems, optimal control of carrier dynamics in nanostructures, and explosion of Xe atomic clusters induced by strong femtosecond laser pulses. As we have shown throughout this work, all these topics are connected with each other and belong to the general topic of properties of electrons in mesoscopic systems at equilibrium and nonequilibrium. In particular, our studies focused on the behavior of electronic systems interacting with external electric fields of various strengths. Our calculations of excitation spectra of few-body quantum systems (see Theory section [2.2] and Results section [3.2]) can be considered in the context of the response of a system interacting with a weak external field, since in this regime the response of the system is fully determined by its spectrum. The optimal control problems considered in this work (see Theory section [2.1],[2.3] and Results section [3.1],[3.3]) belong to the case of weak and moderate fields. In this situation the field does not affect the spectrum of the system significantly. However, one cannot consider the occupations dynamics using perturbation theory, since we are primary interested in significant changes of the occupations. Finally, our investigation of energetic explosion of Xe clusters (see Theory section [2.4] and Results section [3.4]) is an example of the nonperturbative description of the interaction of strong external fields with many-body systems. Since the coupling of electrons with the external field is comparable to the attractive forces inside the Hydrogen atom we treated the coupled dynamics of electrons and ions beyond the adiabatic approximation and beyond the perturbation theory. Let us outline here the most significant results obtained in this work:

• **Optimal control of quantum systems for a finite time interval.** We have presented for the first time a theoretical description of the optimal control for a *finite time interval* in quantum systems with relaxation.

In particular, using a two level system as example, we have shown that relaxation and dephasing effects have a significant influence on the shape of the optimal control fields. A general conclusion which can be drawn on the basis of our calculations is that for a weak relaxation and dephasing in the system the optimal control field consists of pulses more localized in time. In the presence of a moderate relaxation and dephasing in the system the optimal field distributes its intensity (and energy) over the time interval more uniformly avoiding localized peaks. And in the case of a strong relaxation and dephasing a field of a constant amplitude can serve as a good approximation for the optimal field. The same effect takes place for the case of control of the current in a double quantum dot (see Results section [3.3]).

In contrast to other approaches, our theory allows to derive analytical solutions for the optimal control field. This guarantees the uniqueness of the obtained solution, that it is the exact global extremum of the control problem within the model we used. The obtained analytical solutions help to get more physical insight into the optimal control problem. The problems solved analytically in this work can serve as a test for numerical algorithms with the purpose to check their ability to find the optimal solution. Using our theory we found the optimal fields which maximize the time averaged population n_2 of the upper level in two-level systems with and without relaxation. The optimal control problem over a finite time interval represents a nontrivial generalization of known formulations such as proposed by H. Rabitz and co-authors [10] and can provide a more clear interpretation of future experiments in order to perform detailed control of quantum systems. We found that the decoherence processes in the system introduce limits for the optimal control on time interval [0,T], so that T needs to be $T \leq {\gamma_1}^{-1}, {\gamma_2}^{-1}$, where γ_1 and γ_2 are relaxation and dephasing constants, respectively. On a longer time scale one cannot achieve coherent control (see, for example, Fig. [3.25]). In our investigations we account for the relaxation effects phenomenologically by including dissipative terms in the corresponding quantum Liouville equation. In this case we found that decoherence can be only partially compensated by application of the control field. In the considered examples, the optimal fields arising from our approach yield an improvement of a time averaged occupation n_2 of 50% with respect to square or Gaussian pulses of the same energy. We found an absolute upper bound for this kind of optimal control (see Eq. [2.28]). We found that providing more energetic pulses (larger E_0, see Eq. [2.6]) or pulses with a higher time resolution (larger R, see Eq. [2.7]) leads to increase of n_2. Moreover, we have shown that the objective n_2 is a monotonous function of both parameters E_0 and R for the fixed pulse area $\theta(T) = \pi/2$.

Experimentally it is not possible to switch on and switch off the fields at an infinite rate. However, this has not been rigidly enforced in previous

theoretical investigations. To do this we suggested a constraint on the time derivative of the field envelope. By choosing different values of the control parameter R, one can control the minimal width of the optimal pulse. We investigated the influence of the boundary conditions on the optimal control field. In our studies we have considered two cases: the first one is when the amplitude of the control field must be zero at the beginning and at the end of the control interval. The second one is when the control field can be nonzero at the beginning of the control interval. We determine that the field shape is extremely sensitive to the boundary conditions. However, they all have common features like maximal amplitude at the beginning of the control time interval and the resulting evolution of the excited level occupation is similar in both cases. Applications of approximations like the Rotating Wave Approximation or the adiabatic one allowed us to simplify the control problem considerably.

It is interesting to compare our results with other methods where such approximations are not used. In Fig. [4.1] we show the optimal control field obtained by W. Zhu, J. Botina and H. Rabitz [10]. Starting from the occupied ground state of the Morse potential, the authors chose the occupation of the first excited state the value to be maximized at the final time (T=32768 a.u.). This problem can be considered as a limiting case of our theory which we discussed in the Theory section [2.1]. Note, that the field shown in Fig. [4.1] is very close to our solution. The optimal field has a constant amplitude (see Eq. [2.26]). Note, this result we obtain *analytically*. Using the theory developed in this work we also considered

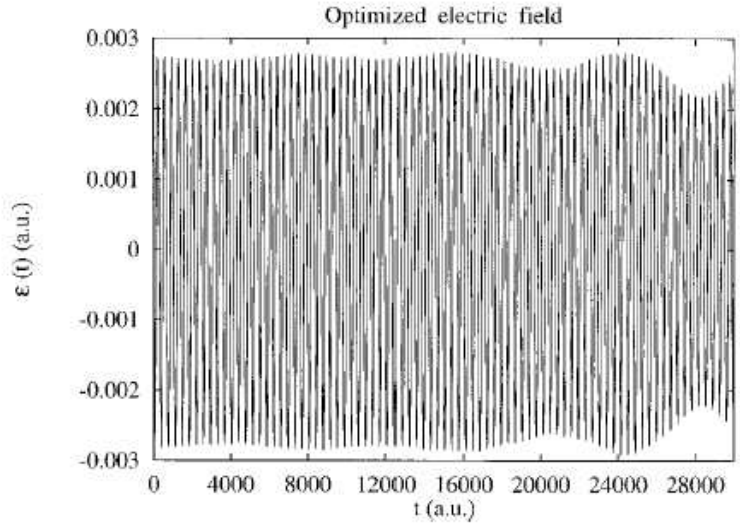

Figure 4.1: Calculated control field in order to maximize the occupation $\rho_{22}(T)$ of the excited level in a two level system at a given time T=32768 a.u. (see [10]).

the optimal control problem concerning the minimization of a time averaged occupation of the excited level n_2 in a two level system (see Theory section [2.1]). We obtained the "inverse hyperbolic cosine" soliton field as a solution for the minimization problem of energy losses during the pulse propagation. Indeed, in the limit of weak decoherence that we have considered the energy losses are proportional to the value of n_2. It will be interesting to generalize this result for the case of larger relaxation and optically dense media. The presented theory is not restricted to the case of one-photon excitations. Using the adiabatic approximation together with Floquet theory, we can generalize our approach in order to treat optimal control problem using multi-photon excitation of the system [70]. We are currently working also on the problem to generalize our analytical approach in order to control a molecular-wave-packet propagation on diabatic surfaces of simple molecules , for instance NaK [5].

Although our understanding of the requirements for optimal control of quantum systems becomes more clear, there remain many fundamental questions that must be addressed. For example, is there a fundamental limit to the control of many-body quantum dynamical processes? How sensitive is the dynamics of controlled quantum systems to fluctuations of the external laser field and other experimental imperfections?

• **Calculation of the excitation spectrum of quantum systems using genetic algorithms.** In this work we also developed a new numerical method - the Quantum Genetic Algorithm (QGA) - to compute the eigenstate problem for quantum systems of few strongly interacting particles. We applied the QGA in order to calculate the eigenstate problem for few electrons confined in a quantum dot realized by various external potentials in one and two dimensions. We first performed calculations in systems for which analytical solutions are known, in order test our method. We found perfect agreement in all cases. Using the QGA we are able to investigate thermal and quantum transitions in few body systems. As a test problem the formation of the "Wigner molecule" in systems of strongly interacting electrons in one and two dimensions was investigated. We also studied the problem of the "Wigner molecule melting" using Hartree-Fock approximation. In our studies we have considered two melting mechanisms, namely, due to thermal and quantum fluctuations. On the base of our results we suppose that the QGA might serve as a basis for the calculation of electronic properties in systems where geometrical effects play a very important role, like quantum billiards.

We should mention that the QGA has also some drawbacks. The main one is that during the procedure one needs to keep in machine memory information about a few hundred possible solutions. This requirement limits the applicability of our method to exact calculations for 3-4 particles only. In order to study larger systems (50-200 particles), it is necessary to use theories like Density Functional Theory (DFT) using various approxima-

tions. The local density approximation, for instance, makes the eigenvalue and eigenstate problem nonlinear due to the nonlinear character of the exchange-correlation functional. In this case most known techniques cannot avoid the problem consisting of being captured in a local minima of the energy functional [89, 111]. In this case the ability of the QGA to find the global extremum and the speed of the convergence exhibited for large nonlinear problems make it very attractive for *ab initio* DFT calculations.

It might be interesting to apply the QGA to the calculation of quantum statistical properties of glassy systems. Since spin glasses exhibit many local minima for the ground state energy functional, the problem to find thermodynamically correct ground state becomes a difficult task [111]. Genetic algorithms can provide a rapid and effective search for the ground state in such systems. With the help of the QGA it is also possible to compute magnetic ground-state properties of magnetic quasi-two-dimensional systems (quantum rings [112]). Since the interaction of electrons in such systems depends on the form of the ring, topological effects should play a significant role. Using genetic procedure to determine the ground state of the system, incorporated into the main GA routine in order to search for the optimal shape of the ring, we plan to find the shape that maximizes the magnetic moment of the system of few interacting electrons [73].

• **Photon assisted tunneling between quantum dots: optimal control approach.** We formulated and solved a problem of the optimal control of the carrier dynamics in nanostructures. As a model device we considered a double quantum dot coupled to metallic contacts and configured as electron pump that was proposed [95] and fabricated recently [115]. The device can be described effectively as a two level system with relaxation. The control problem is formulated with the purpose to maximize the transferred charge through the system. We found that even in the case of moderate decoherence, induced by the coupling to the contacts, it is still possible to perform optimal control in the system and the optimal fields yield up to 80% higher value of the transferred charge with respect to Gaussian or rectangular pulses of the same energy. The results obtained for such a simple model can be useful in the emerging field like quantum computing. Researchers consider quantum dots as the elementary basis for future applications in quantum computing. Since the coherence of electronic states is essential for any operation with quantum bits, it is necessary to know how relaxation effects and decoherence due to the interaction with environment and measurement devices affect the optimal control in nanostructures. This study also has a direct connection with the first topic of our investigations, namely with detailed control of a quantum system over a finite time interval and the effects of decoherence on the optimal control.

In future investigations we plan to consider the control problem in more complex nanoscale objects, for instance, the control of Josephson

junctions [114] and Bose-Einstein condensates [116]. The corresponding equations describing such systems are nonlinear. Under certain conditions it is necessary to perform optimal control in the chaotic regime, where the system dynamics becomes extremely sensitive to control field fluctuations.
• **Explosion of noble gas clusters in strong laser fields.** We have developed a theoretical model to describe the interaction of intense femtosecond laser pulses with Xe clusters. Our results show that the apparent contradictions between the experimental results of Refs. [16] and [17] attributed to different explosion (Coulomb and hydrodynamic) mechanisms do not really exist and that both can be explained from the same phenomenon, namely the inhomogeneous charge distribution caused by the collective electron dynamics. We found that the contribution of the hydrodynamic mechanism does not exceed 20% for the kinetic energy of ejected ions. We suggested a scenario where a cluster becomes inhomogeneously ionized and the electrons are mostly removed from the surface of the cluster. In our calculations we obtain that ejected ions exhibit relatively higher charge state and in the same time lower values of the kinetic energy because of our 1D model. However, our one-dimensional treatment of the clusters contains already, at least qualitatively, all important features of the charge and kinetic-energy distribution of an exploding cluster interacting with an intense laser field. We found that the interaction with intensive fields leads to ultrafast ionization of the cluster. Ionization occurs mainly during the first 20-60 fs corresponding to approximately 5-15 optical cycles, while explosion occurs on relatively longer time scales of 50-200 fs. We have found that the mean energy of the ions after explosion grows with cluster size. However, at sizes more than 10 nm ($\approx$25 atoms in one dimensional "cluster") it exhibits saturation effect that is in qualitative agreement with experimental data and other theoretical models [18].

We plan as a future problem we plan to combine simulation of the cluster explosion with optimization algorithm (based on GA) in order to find the optimal pulse shape that maximizes the total ionization of the fragments or their final kinetic energy. Another project dedicated to the combination of optimal control and intense-laser-pulse matter interaction is a problem to maximize high-harmonics generation emitted by a strongly excited atomic gas using ultrashort laser pulses. It was found recently, that by optimization of the field shape and its frequency modulation it is possible to increase the soft X-ray yield by an order of magnitude [108]. This fascinating result is a nice example of how coherent control can lead to much more effective absorption and emission of photons. With the help of evolutionary algorithms we plan to search for optimal pulses in order to understand the underling physics of the process. Using the Time Dependent formulation of the Density Functional Theory (TDDFT) we plan to simulate the excitation and emission processes.