Abstract

Here two approximate schemes for the description of the interaction between many particle systems and a time varying electromagnetic field are developed. Both methods are based on density functional theory (DFT), which has found widespread use in the determination of ground state properties of quantum mechanical systems. By means of a recently found extension to DFT for time dependent processes, exited states can now also be calculated. The object of this work is the approximate solution of the emerging equations in order to increase the domain of applicability.

For weak external field strengths the so-called γ-approximation has been derived in the context of time dependent perturbation theory. With this parameter free method excitation energies and absorption spectra of large molecules and clusters have been calculated. Benchmark calculations on a set of organic compounds show good agreement with those from more rigorous and time-consuming *first principle* methods and also with the experimentally determined excitations energies and absorption spectra. In addition the optical spectrum of C_{60} is calculated to study the influence of collective effects and their consideration within the γ-approximation.

In order to describe the interaction of ultrashort intense laser pulses with matter, the so-called TD-DFTB scheme is derived in the second part of the thesis. This non-perturbative method is used to realisticly simulate the molecular dynamics on coupled potential surfaces. The focus of the applications lies on the selective excitation of coherent vibrations due to femtosecond laser pulses. Special selection rules found in recent pump-probe experiments have thereby been confirmed on the basis of theoretical calculations. Moreover the simulations yield information on the dependence of competing excitation mechanisms on experimental parameters, for example, pulse duration and laser intensity. Hence they allow to predict future measurements.