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Frank Grossmann

Theoretical Femtosecond Physics

Atoms and Molecules in Strong Laser Fields

Second Edition

Springer

Frank Grossmann
Institut für Theoretische Physik
Technische Universität Dresden
Dresden, Sachsen, Germany

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To the memory of my father
Hans Grossmann

Preface to the Second Edition

The main motivation for this second edition of the book was the inclusion of more exercises and, most importantly, also a collection of solutions of these exercises at the end of the book. Some type errors and more severe flaws of the first edition that have come to my attention have been corrected. The book is, however, intended as a comprehensive introduction to a topic of current interest and not as a complete review of the field. I have therefore not added more (advanced) material into the main text. Furthermore, I have reservedly updated the literature list by including a few more research papers, although a lot more work has been done in the last five years and a related new field, the field of attosecond physics, has emerged. Some more textbook references and reviews have also been added where appropriate.

Again, I would like to express my thanks for enlightening discussions on the topics touched herein to more generations of students that attended my related lectures at TU Dresden and also to Larry Schulman, Jan Handt, Werner Koch, Christoph-Marian Goletz, Alexander Kästner, Michael Fischer, Carlos Zagoya, Sebastian Krause, Niklas Rohling, Max Buchholz, and Tobias Fiedelschuster for ongoing exchange of ideas and to Alexander Kästner for providing numerical results.

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Dresden, Germany — Frank Grossmann
March 2013

Preface to the First Edition

The development of modern pulsed lasers with power densities larger than 10^{16} W/cm^2 and with very short pulse duration in the femtosecond regime enables experimentalists to study elementary processes such as chemical reactions and excitation mechanisms in different areas in physics in the time domain. In parallel to the experimental investigations, analytical and numerical studies of laser driven atoms and molecules with a limited number of degrees of freedom are performed. These theoretical investigations have led to the prediction and/or the explanation of a large variety of partly counter-intuitive phenomena. Among those are the generation of high harmonics using laser excited atoms or molecules, the ionization of atoms above the continuum threshold, the stabilization of atoms against ionization in very strong fields, counter-intuitive pulse sequences to selectively populate vibrational states in molecules and, last but not least, the control of chemical reactions by specially tailored laser pulses.

This book originated out of a course, that I have given on a regular basis since 2000 for advanced undergraduate and graduate students at Technische Universität Dresden. It offers a theoretically oriented approach to the field of laser driven atomic and molecular systems and requires some knowledge of basic classical and quantum mechanics courses as well as of classical electrodynamics. The book has two introductory chapters in Part I that pave the way for the applications in Part II. Part I and also Chap. 3 of Part II contain a selection of textbook material that is needed to understand the rest of the book. The material presented in the last two chapters is close to the recent literature. I have chosen only such works, however, that deal with fundamental concepts and are based on simple model calculations. A biased and incomplete list a references is given at the end of the chapters, preceded by some notes and hints for further reading. For those readers who are interested in some computational details, these are given in the appendices at the end of the corresponding chapters. Furthermore, at several places throughout the text, exercises are placed, whose independent solution allows a deeper understanding of the material presented.

In Chap. 1, we start with a short introduction into the foundations of the laser. We will especially concentrate on those aspects of pulsed lasers that will be important for the theoretical investigations in Part II of the book.

The next fundamental chapter is devoted to the non-relativistic time-dependent Schrödinger equation. In the case of lasers of up to atomic field strengths, this equation allows the theoretical description of the phenomena we want to investigate in Part II. Analytical as well as numerical methods to solve the time-dependent Schrödinger equation are thus in the focus of Chap. 2. Throughout the whole presentation, in order to keep the approach as simple as possible, we touch the topic of correlated many particle dynamics only where necessary and concentrate on the description of electronic as well as nuclear dynamics with the help of models with as few degrees of freedom as possible. The contents and the presentation of Chap. 2 are inspired by the excellent new textbook by David Tannor, *Introduction to Quantum Mechanics: A Time-Dependent Perspective*, which hopefully will start a "revolution" in the way quantum mechanics is taught in the future.

The second part of the book, starting with Chap. 3, contains a collection of equivalent ways to couple a charged particle to a classical electromagnetic field. As the basic postulate we use the principle of minimal coupling. By using unitary transformations, one can then either derive the length form or the Kramers-Henneberger form of the coupling. As first examples of laser-matter interaction, we study the dynamics of (structure-less) two-level systems in laser fields. Phenomena like Rabi oscillations of the occupation probability, occurring there, will be encountered off an on in the remainder of the book. Furthermore, also the fundamental so-called rotating wave approximation will be discussed for the first time in this context.

Selected examples of laser-matter interaction in atomic physics are reviewed in Chap. 4. Here, we concentrate on the phenomena of ionization and high harmonic generation of a single electron in a Coulomb potential of possibly reduced dimensionality. It turns out that a perturbation theoretic approach would not be suited to understand most of the phenomena presented in this chapter. Thus the numerical wavepacket methods that were in the focus of Chap. 2 will find their first application.

The next step in the direction of higher complexity of the dynamics will be taken in Chap. 5. Here we deal with laser-driven systems in molecular and chemical physics. The simplest molecule, the hydrogen molecular ion, H_2^+, will serve as a vehicle to understand some of the basic concepts of molecular physics such as electronic potential surfaces. In the following, the full numerical solution of the coupled electron nuclear problem of H_2^+ in a monochromatic laser field will be reviewed. After discussing the fundamental Born-Oppenheimer approximation, for the rest of the chapter, we then assume that the solution of the electronic many body problem is at our disposal in the form of analytically or numerically given potential energy surfaces. After a short digression on nuclear motion on a single electronic surface, and the discussion of a simple two coupled surfaces problem, we then review some modern applications in the fields of femtosecond spectroscopy, optimal control theory, and quantum information processing under the foregoing assumption.

At this point I thank the students at TU Dresden who have attended my lectures. They have inspired me enormously, through their intense collaboration, during the lectures, as well as during the exercise classes. This has motivated me to rethink

the material presented again and again and the students have thus contributed substantially to the improvement of the manuscript. Also the hospitality of the Max-Planck-Institute for the Physics of Complex Systems, that offered me the opportunity to attend and run several conferences in the field was very important to shape my understanding presented here. Furthermore, I express my deep gratitude to Jan-Michael Rost and Rüdiger Schmidt for their continuous availability for discussions and for long-term collaboration. Moreover, I am grateful to Peter Hänggi for the introduction to the field of driven quantum systems during my Ph.D. work with him, and to Eric Heller for opening the world of time-dependent semiclassics to me. In addition, I have benefitted from uncountable discussions with and valuable advice of former members of the Theoretical Quantum Dynamics Group in Freiburg, especially Gernot Alber, Richard Dehnen, Volker Engel, Christoph Meier, Gerd van de Sand, and Gerhard Stock. Furthermore, former and present members of the Theoretical Atomic and Molecular Physics Group at the Institute of Theoretical Physics of TU Dresden and the Finite Systems Department at the MPIPKS in Dresden have helped shape my understanding. Among many others these are Andreas Becker, Agapi Emmanouilidou, Celal Harabati, Anatole Kenfack, Thomas Kunert, Ulf Saalmann, and Mathias Uhlmann. For helping me by answering specific questions or supplying information and valuable graphs, I would like to thank Wolfgang Schleich, Jan Werschnik, Matthias Wollenhaupt and Shuhei Yoshida. For advice and help with respect to graphics issues, I thank Arnd Bäcker and Werner Koch. Finally, I am indebted to David Tannor, who supplied me with preliminary versions of his book at a very early stage and thus helped shape the presentation here to a substantial degree. The focus of David's book on a time-dependent view of quantum phenomena is an absolute necessity if one wants to study laser-driven systems.

Dresden, Germany Frank Grossmann
May 2008

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