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Theoretical Femtosecond Physics: Atoms and Molecules in Strong Laser Fields

理论飞秒物理
——强激光场中的原子和分子

(影印版)

〔德〕格罗斯曼(F. Grossmann)著

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序　言

物理学是研究物质、能量以及它们之间相互作用的科学。她不仅是化学、生命、材料、信息、能源和环境等相关学科的基础，同时还是许多新兴学科和交叉学科的前沿。在科技发展日新月异和国际竞争日趋激烈的今天，物理学不仅囿于基础科学和技术应用研究的范畴，而且在社会发展与人类进步的历史进程中发挥着越来越关键的作用。

我们欣喜地看到，改革开放三十多年来，随着中国政治、经济、教育、文化等领域各项事业的持续稳定发展，我国物理学取得了跨越式的进步，做出了很多为世界瞩目的研究成果。今日的中国物理正在经历一个历史上少有的黄金时代。

在我国物理学科快速发展的背景下，近年来物理学相关书籍也呈现百花齐放的良好态势，在知识传承、学术交流、人才培养等方面发挥着无可替代的作用。从另一方面看，尽管国内各出版社相继推出了一些质量很高的物理教材和图书，但系统总结物理学各门类知识和发展，深入浅出地介绍其与现代科学技术之间的渊源，并针对不同层次的读者提供有价值的教材和研究参考，仍是我国科学传播与出版界面临的一个极富挑战性的课题。

为有力推动我国物理学研究、加快相关学科的建设与发展，特别是展现近年来中国物理学者的研究水平和成果，北京大学出版社在国家出版基金的支持下推出了“中外物理学精品书系”，试图对以上难题进行大胆的尝试和探索。该书系编委会集结了数十位来自内地和香港顶尖高校及科研院所的知名专家学者。他们都是目前该领域十分活跃的专家，确保了整套丛书的权威性和前瞻性。

这套书系内容丰富，涵盖面广，可读性强，其中既有对我国传统物理学发展的梳理和总结，也有对正在蓬勃发展的物理学前沿的全面展示；既引进和介绍了世界物理学研究的发展动态，也面向国际主流领域传播中国物理的优秀专著。可以说，“中外物理学精品书系”力图完整呈现近现代世界和中国物理

科学发展的全貌，是一部目前国内为数不多的兼具学术价值和阅读乐趣的经典物理丛书。

“中外物理学精品书系”另一个突出特点是，在把西方物理的精华要义“请进来”的同时，也将我国近现代物理的优秀成果“送出去”。物理学科在世界范围内的重要性不言而喻，引进和翻译世界物理的经典著作和前沿动态，可以满足当前国内物理教学和科研工作的迫切需求。另一方面，改革开放几十年来，我国的物理学研究取得了长足发展，一大批具有较高学术价值的著作相继问世。这套丛书首次将一些中国物理学者的优秀论著以英文版的形式直接推向国际相关研究的主流领域，使世界对中国物理学的过去和现状有更多的深入了解，不仅充分展示出中国物理学研究和积累的“硬实力”，也向世界主动传播我国科技文化领域不断创新的“软实力”，对全面提升中国科学、教育和文化领域的国际形象起到重要的促进作用。

值得一提的是，“中外物理学精品书系”还对中国近现代物理学科的经典著作进行了全面收录。20世纪以来，中国物理界诞生了很多经典作品，但当时大都分散出版，如今很多代表性的作品已经淹没在浩瀚的图书海洋中，读者们对这些论著也都是“只闻其声，未见其真”。该书系的编者们在这方面下了很大工夫，对中国物理学科不同时期、不同分支的经典著作进行了系统的整理和收录。这项工作具有非常重要的学术意义和社会价值，不仅可以很好地保护和传承我国物理学的经典文献，充分发挥其应有的传世育人的作用，更能使广大物理学人和青年学子切身体会我国物理学研究的发展脉络和优良传统，真正领悟到老一辈科学家严谨求实、追求卓越、博大精深的治学之美。

温家宝总理在2006年中国科学技术大会上指出，“加强基础研究是提升国家创新能力、积累智力资本的重要途径，是我国跻身世界科技强国的必要条件”。中国的发展在于创新，而基础研究正是一切创新的根本和源泉。我相信，这套“中外物理学精品书系”的出版，不仅可以使所有热爱和研究物理学的人们从中获取思维的启迪、智力的挑战和阅读的乐趣，也将进一步推动其他相关基础科学更好更快地发展，为我国今后的科技创新和社会进步做出应有的贡献。

“中外物理学精品书系”编委会 主任

中国科学院院士，北京大学教授

王恩哥

2010年5月于燕园

F. Grossmann

Theoretical Femtosecond Physics

Atoms and Molecules in Strong Laser Fields

With 91 Figures

Springer

To the memory of my father
Hans Grossmann

Preface

The development of modern pulsed lasers with power densities larger than $10^{16}\,\mathrm{W\,cm^{-2}}$ 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 from a course, that I have been giving 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 of textbook knowledge 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 of 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 concentrate especially 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 book, 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 and 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, in 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 consider the material presented here 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 PhD work with him, and to Eric Heller for opening the world of time-dependent semiclassics to me. In addition, I have benefitted from innumerable 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 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, May 2008 *Frank Grossmann*

Contents

Part I Prerequisites

Part I

Prerequisites

1

A Short Introduction to Laser Physics

To study the influence of light on the dynamics of an atom or a molecule experimentally, laser light sources are used most frequently. This is due to the fact that lasers have well-defined properties. The theory of the laser dates back to the 1950s and 1960s of the twentieth century and by now, 50 years later is textbook material. In this introductory chapter, we start by recapitulating some basic notions of laser theory, which will be needed to understand later chapters.

More recently, experimentalists have been focusing on pulsed mode operation of lasers with pulse lengths of the order of femtoseconds, allowing for time-resolved measurements. At the end of this chapter, we therefore put together some aspects of pulsed lasers that are important for their application to atomic and molecular systems.

1.1 The Einstein Coefficients

Laser activity may occur in the case of nonequilibrium, as we will see later. Before dealing with this situation, let us start by considering the case of equilibrium between the radiation field and an ensemble of atoms in the walls of a cavity. This will lead to the Einstein derivation of Planck's radiation law.

The atoms will be described in the framework of Bohr's model of the atom, allowing the electron to occupy only discrete energy levels. For the derivation of the radiation law, the consideration of just two of those levels is sufficient. They shall be indexed by 1 and 2 and shall be populated such that for the total number of atoms

$$N = N_1 + N_2 \tag{1.1}$$

holds. This means that N_2 of the atoms are in the excited state with energy E_2 and N_1 atoms are in the ground state with energy E_1. Transitions between the states shall be possible by emission or absorption of photons of the appropriate energy. The following processes can be distinguished: