Attosecond and Strong-Field Physics

Principles and Applications

Probing and controlling electrons and nuclei in matter at the attosecond timescale became possible with the generation of attosecond pulses by few-cycle intense lasers, revolutionizing our understanding of atomic structure and molecular processes. This book provides an intuitive approach to this emerging field, utilizing simplified models to develop a clear understanding of how matter interacts with attosecond pulses of light. An introductory chapter outlines the structures of atoms and molecules and the properties of a focused laser beam. Detailed discussion of the fundamental theory of attosecond and strong-field physics follows, including the molecular tunneling ionization model, the quantitative rescattering model, and the laser-induced electronic diffraction theory for probing the change of atomic configurations in a molecule. Highlighting cutting-edge developments in attosecond and strong-field physics, and identifying future opportunities and challenges, this self-contained text is invaluable for students and researchers in the field.

C. D. Lin is a University Distinguished Professor at Kansas State University. His research group has made important contributions to the field of attosecond science, including the development of the molecular tunneling ionization model and the quantitative rescattering model.

Anh-Thu Le is a research professor at Kansas State University. For more than twenty years, he has studied atomic, molecular, and optical physics, and, with C. D. Lin, developed the quantitative rescattering theory for high-order harmonic generation.

Cheng Jin is a professor at Nanjing University of Science and Technology who studies optimization of the generation of isolated attosecond pulses by synthesis of multicolor laser waveforms in the gas medium.

Hui Wei is a postdoctoral fellow at Kansas State University. His research interests include characterization and applications of attosecond pulses to molecules and solids.

"This is the book we were waiting for. The text covers strong field light-matter interaction and its effects for atoms and molecules in a concise but easy to read manner and serves as an excellent reference. Topics range from the fundamentals of ionization, the effects of molecular symmetry, high harmonic and attosecond pulse generation, ATI and LIED to attosecond dynamics in atoms and molecules. This book is equally useful for the experienced research as it is for a senior undergraduate student in physics of chemistry. Definitely a must-have!"

Professor Jens Biegert, The Institute of Photonic Sciences (ICFO)

"The book by Lin et al. provides a very accessible introduction to the emerging fields of attosecond science and strong field laser physics, combining a textbook-style presentation of the underlying physics with a presentation of selected recent research results. This book will be a valuable resource for both new students and experienced researchers in the field."

Professor Mark Vrakking, Max Born Institute, Berlin

"Attosecond science based on strong field physics are written comprehensively, including generation, measurement and application of attosecond pulse. This book can be not only used as a textbook for graduate students but also useful for researchers as the first reference in attosecond and strong field community."

Dr Katsumi Midorikawa, Director, RIKEN Center for Advanced Photonics

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C. D. LIN
Kansas State University

ANH-THU LE
Kansas State University

CHENG JIN
Nanjing University of Science and Technology

HUI WEI
Kansas State University

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UNIVERSITY PRESS

CAMBRIDGE
UNIVERSITY PRESS

University Printing House, Cambridge CB2 8BS, United Kingdom

One Liberty Plaza, 20th Floor, New York, NY 10006, USA

477 Williamstown Road, Port Melbourne, VIC 3207, Australia

314–321, 3rd Floor, Plot 3, Splendor Forum, Jasola District Centre, New Delhi – 110025, India

79 Anson Road, #06–04/06, Singapore 079906

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Preface

The observation of second-harmonic generation by Franken and coworkers back in 1961 preceded 40 years of fast growth in the field of nonlinear optics as well as the first demonstration of the generation of attosecond pulses in 2001. Following the first report of high-order harmonic generation in rare gas atoms with intense-focused laser beams in 1987 and the subsequent emergence of titanium-sapphire lasers, it became clear that the generated high-order harmonics are phase locked. Much like how earlier mode-locked lasers led to femtosecond laser pulses, these phase-locked high-order harmonics were expected to lead to attosecond pulses. Still, fine-tuning the technique utilized to successfully measure the phases of harmonics took more than ten years of effort by pioneers in strong-field physics. During this time, laser-atom/molecule interactions and the properties of harmonics were widely studied both theoretically and in laboratories. Thus, the birth of attosecond science at the dawn of the twenty-first century owes much to the advance of strong-field physics in the decade before.

Today, attosecond pulses ranging from vacuum ultraviolet (VUV) to soft X-rays are available for applications, especially on atoms and molecules. Much of what has been understood in this field resulted from new experimental detection of electrons and ions together with high-resolution spectra borrowed from conventional atomic and molecular physics. The last fifteen years have witnessed the rapid growth of the field and that pace is accelerating.

Researchers now entering this field come with diverse backgrounds and expertise in physics, optics, chemistry, or engineering. This diversity speaks to the richness and potential of attosecond science that has found applications in systems ranging from simple atoms to complex molecules, nanostructures, and condensed materials. Attosecond pulses have been promised to probe electron dynamics in the attosecond timescale, but what parameters should be measured to characterize the dynamics? Can the theoretical tools and concepts developed for attosecond science and strong-field physics be scaled up to complex systems where the knowledge of the target is very limited?

The rapid developments in attosecond science in recent years have been widely reviewed in a number of journal articles and monographs. These publications tend to focus on specific knowledge obtained from experimental findings. In this book, we try to treat attosecond and strong-field physics as a cohesive subject. We examine the underlying physics behind the simple models used in this field. In addition, concepts used for qualitative understanding are distinguished from theoretical models that can make approximate predictions on experimental outcomes. Clear distinction of the roles of the concept and the theoretical model is essential in order to avoid the confusion that can lead to nonconstructive debates.

Attosecond physics forces practitioners away from the comfort zone of conventional energy-domain physics. Asking the right questions is an important part of this endeavor and this book does not shy away from addressing controversial issues such as the tomographic imaging of molecular orbitals, the photoionization time delay, and the description of charge migration or hole-hopping dynamics.

This book has been written with beginning graduate students in mind and can be used as a textbook for a two-semester course on strong-field and attosecond physics. Instructors may also select individual chapters for a one-semester course and serious students are advised to complete the exercises and fill out the missing steps in the derivations. Furthermore, we hope that this book will be used as a reference in the attosecond and strong-laser-field community.

We begin Chapter 1 with a brief review of atoms, molecules, and wave propagation. For atomic physics, wavefunctions of bound states and continuum states within the one-electron model are considered. Simple scattering theory and single-photon ionization theory are then presented. We then review helium atoms (including Fano resonances), many-electron atoms at the shell model level, and the simple density functional theory. For molecules, we discuss the rotational, vibrational, and electronic wavefunctions and spectroscopy for diatomic and polyatomic molecules. Furthermore, elementary group theory for describing polyatomic molecules is introduced to help readers understand the classification of molecular states. Finally, the propagation of a focused laser beam in space and through optical elements such as lenses or mirrors is treated.

Chapter 2 is devoted to the formal theories used in strong-field physics. Topics include weak-field and strong-field expansion methods, tunnel ionization theory, the Keldysh-Faisal-Reiss (KFR) theory and its various extensions, and the classical trajectory Monte Carlo (CMTC) method and its extension. These approximate theories serve as a basis for understanding the strong-field experiments treated in the following chapters.

Chapter 3 is devoted to ionization of atoms by an intense laser field. We cover total ionization, excitation, energy, and momentum distributions of photoelectrons and the surprising features of very low-energy electrons generated by mid-infrared lasers. Chapter 3 also discusses the theory of orientation and alignment of molecules and of creating vibrational wave packets.

In Chapter 4, rescattering phenomena are described. Beginning with the simple three-step picture, this concept is generalized to the quantitative rescattering (QRS) theory. The QRS model was first properly calibrated against results from solving the time-dependent Schrödinger equation. It is shown that high-energy photoelectron angular distributions can be used to extract elastic, differential-scattering cross-sections. This theory is then applied to laser-induced electron diffraction (LIED), where the bond lengths of a molecule can be extracted. Realization of LIED for small molecules in the laboratory are also illustrated in this chapter.

Chapters 5 and 6 are devoted to high-order harmonic generation. Harmonics generated within a single atom picture are first discussed in Chapter 5, using strong-field approximation, quantum orbits theory, and QRS theory. Then, descriptions of macroscopic propagation of the driving laser and harmonics in the gas medium are given. Chapter 5 closes by comparing experimental high-harmonic generation (HHG) spectra for rare gas atoms to QRS-based HHG theory including propagation effect. Applications of the QRS theory to HHG spectra from diatomic and polyatomic molecules are treated in Chapter 6. Here it is shown that photoionization dipole-transition matrix elements (including amplitude and phase) can be extracted from the HHG spectra under favorable conditions. Chapter 6 ends with a summary of recent efforts around the enhancement of harmonic yields and the extension of harmonics to the water-window region.

Chapters 7 and 8 are dedicated to attosecond physics. After a short discussion on the attosecond pulse train, we summarize the various methods of the generation of isolated attosecond pulses (IAP). Then, the various methods of the metrology of IAP are discussed. Chapter 7 concludes with a detailed analysis of the photoionization time delay. Attosecond pulses as a tool for probing electron dynamics are treated in Chapter 8. Starting with a description of Fano resonance in the time domain, Chapter 8 shows that an "electronic movie" (as depicted on the cover of this book) was recently filmed using the method of attosecond transient absorption spectroscopy (ATAS). Because it offers extreme temporal and spectral resolution, ATAS is a powerful method for attosecond sciences. The potential power of attosecond pulses for probing electron dynamics is also addressed in Chapter 8.

The choice of materials in this book is biased toward the expertise of the authors. It cannot cover all aspects of the recent important advances in attosecond and strong-field physics. Thus, for instance, only interactions with linearly polarized laser pulses are included. In addition, we do not feel comfortable covering recent developments in applications of attosecond pulses and strong fields on solid materials, nanostructures, and surfaces. A separate book by experts in this area is certainly necessary.

Many graduate students and postdoctoral scholars contributed to the materials covered in this book. They are recognized here: Zhangjin Chen, Wei-Chun Chu, Sam Micheau, Toru Morishita, Xiao-Min Tong, Xu Wang, Junliang Xu, Xi Zhao, and Zengxiu Zhao. Visiting scholars have also contributed to the topics covered in this book. These include Zigen Chen, Van-Hoang Le, Qianguang Li, Van-Hung Hoang, Shicheng Jiang, Ty Nguyen, Guoli Wang, M. Wickenhauser, Yan Wu, Shan Xue, Chao Yu, Song-Feng Zhao, Xiao-Xin Zhou, and Zhaoyan Zhou. Moreover, the time-dependent Schrödinger equation (TDSE) code from Toru Morishita and the molecular photoionization code from Robert Lucchese were undeniably essential to the success of the QRS theory.

Fruitful collaborations with experimental groups at Kansas State University and elsewhere on topics covered in this book should also be mentioned, in particular the groups of Ravi Bhardwaj, Jens Biegert, Ming-Chang Chen, Lew Cocke, Paul Corkum, Lou DiMauro and Pierre Agostini, Kyung-Han Hong, Andy Kung, Thomas Pfeifer, Kiyoshi Ueda, and David Villeneuve.

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