

# Preface

The properties of matter are ultimately determined by its electronic structure. A suitable perturbation of the electrons' distribution from its equilibrium configuration in a system such as an atom, a molecule, or a solid, can therefore initiate certain dynamical processes. Intense and ultrashort light pulses are ideal tools for that purpose as their electric fields couple directly to the electrons. Therefore, they are not only able to distort the equilibrium electron distribution but, even allow driving a system on sub-femtosecond timescales with the light's Petahertz field-oscillations. This has been realized first in experiments that studied atoms in strong laser fields some 25 years ago. These experiments revealed a wealth of fundamentally important phenomena that are strictly timed to the laser-field oscillations such as the release of electrons by tunneling through the field-distorted Coulomb binding potential, or field-driven (re-)collisions of the released electrons with the atomic ion. The latter, in turn, led to the discovery of a range of essential secondary processes such as the generation of very high orders of harmonics of the driving light with photon energies that can extend into the X-ray range.

Since then, thanks to a true revolution in laser technology, tremendous progress has been made in the field. Laser science has now reached a level of perfection where it is possible to produce intense light pulses with durations down to a single oscillation cycle and with virtually arbitrary evolution of the electric field in a wide range of frequencies. The availability of such field transients enabled a number of exciting possibilities, such as control over the breakage of selected chemical bonds in molecules by directly driving the molecular valence electrons that actually form the bond, or the production of coherent attosecond pulses in the soft X-ray wavelength range that can be used for probing or initiating dynamics on time-intervals during which the electronic distribution in the system under study stays essentially frozen.

The recent years have seen a particularly vivid progress in the research of using ultrashort intense light pulses for controlling and probing ultrafast dynamics. On the one hand, a number of groups have extended the research field to systems with a much increased complexity and have studied and controlled field-induced dynamics in large polyatomic molecules, cluster complexes, bio-matter, nanoparticles and crystals, and also in condensed phase systems such as solid surfaces,

nanostructures, and bulk solids. On the other hand, the availability of new, coherent light sources in both the very short (X-rays) and very long (mid-infrared) wavelength ranges have allowed for the production and application of short and intense pulses in previously unexplored regimes.

Owing to this large diversity of pulsed sources and dynamical systems studied with them it becomes inherently difficult to provide a unique definition and sharp boundaries for this field of research. This is also reflected by the considerable variety of titles used for conferences in this field. Obviously the same difficulty arises in providing a clear and well-defined but at the same time comprehensive title for a book on this research field. Although *ultrafast dynamics* is a relative term and covers a large range of dynamical processes and timescales, including rotational and vibrational dynamics, we would like to define this term here as electronic dynamics and processes that result from an essentially instantaneous distortion of the equilibrium electronic structure in a given system.

With this book we have tried, by carefully picking 14 examples of cutting-edge scientific research, grouped in four areas, to provide a comprehensive overview not only over the current state of the research field that uses ultrashort intense light pulses and light sources based on these pulses for initiating, driving, controlling, and probing ultrafast dynamics, but also over its recent tremendous and exciting developments. With the selection of the four areas we have attempted to provide the broadest possible overview over the such defined research field by covering essentially all currently studied physical systems from individual atoms and molecules to nanostructures and bulk macroscopic media, and all available ultrafast pulse sources from the mid-infrared to the X-ray range. Of particular importance for us was to highlight the possibilities that are opened up by the availability of new light sources, and the new research questions that arise by pushing research toward new systems with increased complexity such as nanostructures and bulk macroscopic media. Also, we have tried to provide both an experimental and theoretical perspective on the research field. The book is structured as follows.

*The first part* of research that is discussed during the first four chapters shall provide an overview of the possibilities that a strong laser field opens up for controlling electronic processes in atoms, molecules, nanostructures, and solids. *The second part* of the book is dedicated to the application of intense laser pulses in combination with attosecond pulses, obtained by the laser-driven process of high-harmonic generation, for triggering and probing ultrafast dynamics. *The third part* of the book discusses in four chapters the only very recently opened research route of using ultrashort intense laser pulses for driving electronic dynamics on surfaces, in nanostructures and in solids. While on the one hand this type of research is interesting from a fundamental point of view as it investigates the interaction of light and matter in completely new regimes of parameters where collective effects, material parameters, and system geometries start to play a role, this research also comprises considerable potential for applications in that it could be used for, e.g., fabrication of devices for information transmission or fast switching. A particularly important process in this context is the excitation of collective surface electron oscillations called a surface plasmon. *The fourth and*

*final part* of the book is dedicated to the exciting possibilities that are opened up by the availability of intense light pulses in the X-ray wavelength regime that can be produced by free-electron lasers, the first of which have started their operation just a few years ago. The two chapters in this part discuss applications of such pulses depicted by examples of research performed at the free-electron lasers in Hamburg (FLASH) and Stanford (Linac Coherent Light Source, LCLS), respectively.

We hope that this book will be equally inspiring and helpful for young researchers, who would like to step into this field, and for experienced researchers who may enjoy the exhaustive discussion that covers the research on essentially all currently studied objects and with all available ultrafast pulse sources in the field that uses ultrashort light pulses for controlling and probing ultrafast dynamics.

Vienna  
Jena

Markus Kitzler  
Stefanie Gräfe

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From Atoms to Solids, from Lasers to Intense X-rays

Kitzler, M.; Gräfe, S. (Eds.)

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