Ultrashort Laser Pulse Phenomena

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Fundamentals, Techniques, and Applications on a Femtosecond Time Scale

Second Edition

JEAN-CLAUDE DIELS
Department of Physics and Astronomy
University of New Mexico
Albuquerque, NM

WOLFGANG RUDOLPH
Department of Physics and Astronomy
University of New Mexico
Albuquerque, NM

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Preface

Almost 10 years have passed since the first edition of *Ultrashort Laser Pulse Phenomena*. The field of ultrafast optics and spectroscopy has evolved and matured tremendously; tools and techniques available only in research laboratories 10 years ago are now common in many laboratories outside physics and engineering and have been commercialized. During the same period the field has progressed at an astonishing speed, opening new directions, constantly challenging the frontiers of high field science and ultrafast spectroscopy. Our provocative statement from the first edition predicting attosecond pulses at the end of the 1990s materialized. To properly account for the developments of the past decade each chapter of the first edition would need to be expanded into a whole book.

Having said this it is clear that this second edition, like the first edition, cannot be an attempt to review and summarize the latest developments in the field. Periodic updates can be found in the proceedings of the conferences on ultrafast phenomena and on ultrafast optics held alternately every other year. However, as is typical for a mature scientific area, despite the dramatic progress a number of fundamental subjects have emerged. These topics, not much different from the material covered in the first edition, are what students and researchers entering the field need to learn.

In line with the scope of the first edition, the second edition is also intended to bridge the gap between a textbook and a monograph. Written at the level of senior undergraduate students from physics, chemistry, or engineering it represents a mix of tutorial sections and more advanced writings motivating further study of the original literature.

Compared to the first edition, changes have been made in particular in Chapters 1, 2, 3, 5, 9, and 13. The tutorial aspect was emphasized more, and material useful for the researcher was added. The original Chapter 5 on "Ultrashort Sources" has been expanded and split into two chapters, Chapter 5 on "Fundamentals" and Chapter 6 on "Examples." Some newer developments were added to Chapter 9 on "Diagnostic Techniques" and to Chapter 13 on "Selected Applications." Except for some updates and corrections, Chapters 7, 8, 10, 11, and 12 are essentially unchanged.

We would like to express our gratitude to all our colleagues and students who have supported us with numerous suggestions and corrections. In particular, we are indebted to current and former students L. Arissian, J. Biegert, M. Dennis, S. Diddams, P. Dorn, J. Jasapara, J. Jones, M. Kempe, A. Knorr, M. Mero,

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We are grateful also to the contributions of all the students who took courses in the development stage of the first and second editions of this book and proofread individual sections.

Last but not least, we are grateful to our wives, who watched the years go by as our lives became hostage to this endeavor.

Our apologies again to anyone whose work has not been adequately recognized, as we could not possibly cover completely the macrocosm of the temporal microcosm.

Albuquerque, December 2005

Preface to the First Edition

What do we understand about "ultrashort laser pulse phenomena"? It really takes a whole book to define the term. By ultrashort we mean femtosecond (fs), which is a unit of time equal to 10^{-15} s. This time scale becomes accessible because of progress in the generation, amplification, and measurement of ultrashort light pulses. Ultrashort phenomena involve more than just the study of ultrashort lived events. Because of the large energy concentration in a fs optical pulse, this topic encompasses the study of the interaction of intense laser light with matter, as well as the transient response of atoms and molecules and basic properties of the fs radiation itself.

This book is intended as an introduction to ultrashort phenomena to researchers and graduate and senior undergraduate students in optics, physics, chemistry, and engineering. A preliminary version of this book has been used at the University of New Mexico, Jena and Pavia, as a course for graduate and advanced undergraduate students. The femtosecond light gives a different illumination to some classical problems in electromagnetism, optics, quantum mechanics, and electrical engineering. We believe therefore that this book can provide useful illustrations for instructors in these fields.

It is *not* the goal of this book to represent a complete overview of *the latest* progress in the field. We wish to apologize in advance for all the important and pioneering fs work that we failed to cite. For space limitation, we have chosen to present only a few examples of application in the various fields. We are not offering different theoretical aspects of any particular phenomenon, but rather choose to select a description that is consistent throughout the book. Our aim is to cover the basic techniques and applications rather than enter into details of the most fashionable topic of the day. We have attempted to use simple notations and to remain within the MKS system of units.

Consistent with the instructional goal of this book, the first chapter is an extensive review of propagation properties of light in time and frequency domains. Classical optics is reviewed in the next chapter, in light of the particular propagation properties of fs pulses. Some aspects of white light optics—such as coherence and focusing—can be explained in the simplest manner by picturing incoherent radiation as a random sequence of fs pulses. Femtosecond pulses are generally meant to interact with matter. Therefore, a review of this aspect is given in Chapter 3. The latter serves as introduction to the most startling, unexpected, complex properties of transient interaction of coherent fs pulses with resonant

physical and chemical systems (Chapter 4). This is a subfield of which the basic foundations are well understood, but it is still open to numerous experimental demonstrations and applications. Chapters 5 through 9 review practical aspects of femtosecond physics, such as sources, amplifiers, pulse shapers, diagnostic techniques, and measurement techniques.

The last three chapters are examples of application of ultrafast techniques. In Chapter 10, the frontier between quantum mechanics and classical mechanics is being probed with fs pulses. New techniques make it possible to "visualize" electrons in Rydberg orbits or the motion of atoms in molecules. The examples of ultrafast processes in matter are presented in this chapter by order of increasing system complexity (from the orbiting electron to the biological complex).

Femtosecond pulses of high peak powers lead to the generation of extremely short wavelength electron and X-ray pulses, as well as to extremely long wavelengths. Some of these techniques are reviewed in Chapter 11. On the long wavelength end of the spectrum, fs pulses are used as Dirac delta function on antennas for submillimeter radiation (frequencies in the THz range). This is a recent application of ultrafast solid-state photoconductive switches.

A few applications that exploit the short duration (range gating imaging), the high coherence, or the high intensity (solitons or filamentation in air) have been selected for the final Chapter 13.

Problems are given at the end of most chapters. Some are typical textbook problems with a straightforward solution. Other problems are designed to put the student in a realistic research situation.

Why Ultrashort Pulse Phenomena?

Yes, you are right! You can be happy without femtosecond pulses and, maybe, consider yourself lucky enough not to be involved with it too deeply. Nevertheless it is a fascinating as well as challenging task to observe and to control processes in nature on a time scale of several femtoseconds. Note, one femtosecond (1 fs) is the $1 : 10^{15}$th part of a second and corresponds to about half a period of red light. The ratio of one fs to one second is about the ratio of 5 minutes to the age of the earth. During one fs, visible light travels over a distance of several hundred nanometers, which is hardly of any concern to us in our daily routine. However, this pathlength corresponds to several thousand elementary cells in a solid which is quite a remarkable number of atomic distances. This suggests the importance the fs time scale might have in the microcosm. Indeed, various essential processes in atoms and molecules, as well as interactions among them, proceed faster than what can be resolved on a picosecond time scale ($1\ \text{ps} = 10^{-12}\text{s}$). Their relevance results simply from the fact that

these events are the primary steps for most (macroscopic) reactions in physics, chemistry, and biology.

To illustrate the latter point, let us have a look at the simplest atom—the hydrogen atom—consisting of a positively charged nucleus and a negatively charged electron. Quantum mechanics tells us that an atomic system exists in discrete energy states described by a quantum number n. In the classical picture this corresponds to an electron (wave packet) circulating around the proton on paths with radius $R \propto n^2$. From simple textbook physics, the time T_R necessary for one round-trip can be estimated with $T_R = 4n^2h^3\epsilon_0^2/(e^4m_e)$, where h is Planck's constant, ϵ_0 is the permittivity of free space, and m_e, e are the electron mass and charge [1, 2]. For $n = 26$, for instance, we obtain a period of about 100 fs. Consequently, an (hydrogen) atom excited to a high Rydberg state is expected to show some macroscopic properties changing periodically on a fs time scale.

Let us next consider atoms bound in a molecule. Apart from translation, the isolated molecule has various internal degrees of freedom for periodical motion—rotation and vibration as well as for conformation changes. Depending on the binding forces, potentials, and masses of the constituents, the corresponding periods may range from the ps to the fs scale. Another example of ultrafast dynamics in the molecular world is the chemical reaction, for instance, the simple dissociation $(AB)^* \rightarrow A + B$. Here the breaking of the bond is accompanied by a geometrical separation of the two components caused by a repulsive potential. Typical recoil velocities are of the order of 1 km/s, which implies that the transition from the bound state to the isolated complexes proceeds within 100 fs. Similar time intervals, of course, can be expected if separated particles undergo a chemical reaction.

Additional processes come into play if the particle we look at is not isolated but under the influence of surrounding atoms or molecules, which happens in a gas (mixture) or solution. Strong effects are expected as a result of collisions. Moreover, even a simple translation or rotation that alters the relative position of the molecule to the neighboring particles may lead to a variation of the molecular properties because of a changed local field. The characteristic time constants depend on the particle density and the translation–rotation velocity, which in turn is determined by the temperature and strength of interaction with the neighbors. The characteristic times can be comparatively long in diluted gases (ns to μs) and can be short in solutions at room temperature (fs).

Finally, let us have a look at a solid where the atomic particles are usually trapped at a relatively well-defined position in the lattice. Their motion is restricted usually to lattice vibrations (phonons) with possible periods in the order of 100 fs, which corresponds to phonon energies of several tens of milli-electronvolts (for instance, the longitudinal optical or LO phonon in GaAs has an energy of about 35 meV).

The fundamental problem to be solved is to find tools and techniques that allow us to observe and manipulate on a fs time scale. At present, speaking about tools and techniques for fs physics means dealing with laser physics, in particular with ultrashort light pulses produced in lasers. Shortly after the invention of the laser in 1960, methods were developed to use them for the generation of light pulses. In the sixties, the microsecond (μs) and nanosecond (ns) range were extensively studied. In the seventies, progress in laser physics opened up the ps range, and the eighties were characterized by the broad introduction of fs techniques (extrapolating this dramatic development we may expect the attosecond physics in the late nineties). Optical methods have taken precedence over electronics in time resolving fast events ever since light pulses shorter than a few ps have become available. It should also be mentioned that the shortest electrical and X-ray pulses are now being produced by means of fs light pulses, which in turn enlarges the application field of ultrafast techniques.

Femtosecond technology opens up new fascinating possibilities based on the unique properties of femtosecond light pulses:

- Energy can be concentrated in a temporal interval as short as several 10^{-15} s, which corresponds to only a few optical cycles in the visible range.
- The pulse peak power can be extremely large even at moderate pulse energies. For instance, a 50-fs pulse with an energy of 1 mJ ($\approx 3 * 10^{15}$ "red" photons) exhibits an average power of 20 Gigawatt. Focusing this pulse to a 100-μm^2 spot yields an intensity of 20 Petawatt/cm^2 (20 10^{15} W/cm^2!), which means an electric field strength of about 3 GV/cm. This value is larger than a typical inner-atomic field of 1 GV/cm.
- The geometrical length of a fs pulse amounts only to several micrometers (10 fs corresponds to 3 μm in vacuum). Such a coherence length is usually associated with incoherent light. The essential difference is that incoherent light is generally spread over a much longer distance.

The attractiveness of fs light pulses not only lies in the possibility to trace processes in their ultrafast dynamics, but also in the fact that one simply can do things faster. Of course only a few, but essential, parts in modern technology can be accelerated by using ultrashort (fs) light pulses. Of primary importance are data transfer and data processing utilizing the high carrier frequency of light and the subsequent large possible bandwidths. In this respect one of the most spectacular goals is to create an optical computer. Moreover, techniques are being developed that allow distortionless propagation of ultrashort light pulses over long distances (several thousand kilometers) through optical fibers, a precondition for a future Terahertz information transfer.

A variety of nonlinear processes, reversible as well as irreversible ones, become accessible thanks to the large intensities of fs pulses. There are proposals

to use such pulses for laser fusion. To reach TW intensities, tabletop devices are replacing the building size high energy facilities previously required. First attempts to generate short X-ray pulses by using fs pulse–induced plasmas have already proven successful.

The short geometrical lengths of fs light pulses suggest interesting applications for optical ranging with micrometer resolution, as well as for combinations of micrometer spatial resolution with femtosecond temporal resolution.

The ultrashort phenomena to which this book refers are created by *light pulses*, which are wave packets of electromagnetic waves oscillating at optical frequencies. The emphasis of this book is not on the optical frequency range but on physical phenomena associated with *ultrafast* electromagnetic pulses. The latter will be ephemeral when consisting of only a small number of optical periods and spatially confined when made up of a small number of wavelengths. Another criterion for short is that the length of the pulse be small compared with the distance over which it propagates, particularly when large changes of shape and modulation take place. In the particular area of light–matter interaction, a pulse is generally considered as a δ function excitation when its duration is small compared to that of all atomic or molecular relaxations.

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