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国外物理名著系列 4

（影印版）

Femtosecond Laser Pulses
Principles and Experiments
(2nd Edition)

飞秒激光脉冲
——原理及实验
（第二版）

C. Rullière

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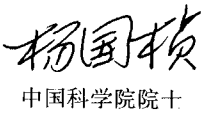
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国外物理名著(影印版)系列序言

对于国内的物理学工作者和青年学生来讲，研读国外优秀的物理学著作是系统掌握物理学知识的一个重要手段。但是，在国内并不能及时、方便地买到国外的图书，且国外图书不菲的价格往往令国内的读者却步，因此，把国外的优秀物理原著引进到国内，让国内的读者能够方便地以较低的价格购买是一项意义深远的工作，将有助于国内物理学工作者和青年学生掌握国际物理学的前沿知识，进而推动我国物理学科研和教学的发展。

为了满足国内读者对国外优秀物理学著作的需求，科学出版社启动了引进国外优秀著作的工作，出版社的这一举措得到了国内物理学界的积极响应和支持，很快成立了专家委员会，开展了选题的推荐和筛选工作，在出版社初选的书单基础上确定了第一批引进的项目，这些图书几乎涉及了近代物理学的所有领域，既有阐述学科基本理论的经典名著，也有反映某一学科专题前沿的专著。在选择图书时，专家委员会遵循了以下原则：基础理论方面的图书强调“经典”，选择了那些经得起时间检验、对物理学的发展产生重要影响、现在还不“过时”的著作(如：狄拉克的《量子力学原理》)。反映物理学某一领域进展的著作强调“前沿”和“热点”，根据国内物理学研究发展的实际情况，选择了能够体现相关学科最新进展，对有关方向的科研人员和研究生有重要参考价值的图书。这些图书都是最新版的，多数图书都是2000年以后出版的，还有相当一部分是2006年出版的新书。因此，这套丛书具有权威性、前瞻性和应用性强的特点。由于国外出版社的要求，科学出版社对部分图书进行了少量的翻译和注释（主要是目录标题和练习题)，但这并不会影响图书“原汁原味”的感觉，可能还会方便国内读者的阅读和理解。

“他山之石，可以攻玉”，希望这套丛书的出版能够为国内物理学工作者和青年学生的工作和学习提供参考，也希望国内更多专家参与到这一工作中来，推荐更多的好书。



中国科学院院士
中国物理学会理事长
2007年3月20日

Preface

This is the second edition of this advanced textbook written for scientists who require further training in femtosecond science. Four years after publication of the first edition, femtosecond science has overcome new challenges and new application fields have become mature. It is necessary to take into account these new developments. Two main topics merged during this period that support important scientific activities: attosecond pulses are now generated in the X-UV spectral domain, and coherent control of chemical events is now possible by tailoring the shape of femtosecond pulses. To update this advanced textbook, it was necessary to introduce these fields; two new chapters are in this second edition: "Coherent Control in Atoms, Molecules, and Solids" (Chap. 11) and "Attosecond Pulses" (Chap. 12) with well-documented references.

Some changes, addenda, and new references are introduced in the first edition's ten original chapters to take into account new developments and update this advanced textbook which is the result of a scientific adventure that started in 1991. At that time, the French Ministry of Education decided that, in view of the growing importance of ultrashort laser pulses for the national scientific community, a Femtosecond Centre should be created in France and devoted to the further education of scientists who use femtosecond pulses as a research tool and who are not specialists in lasers or even in optics.

After proposals from different institutions, Université Bordeaux I and our laboratory were finally selected to ensure the success of this new centre. Since the scientists involved were located throughout France, it was decided that the training courses should be concentrated into a short period of at least 5 days. It is certainly a challenge to give a good grounding in the science of femtosecond pulses in such a short period to scientists who do not necessarily have the required scientific background and are in some cases involved only as users of these pulses as a tool. To start, we contacted well-known specialists from the French femtosecond community; we are very thankful that they showed enthusiasm and immediately started work on this fascinating project.

Our adventure began in 1992 and each year since, generally in spring, we have organized a one-week femtosecond training course at the Bordeaux University. Each morning of the course is devoted to theoretical lectures concerning different aspects of femtosecond pulses; the afternoons are spent in the laboratory, where a very simple experimental demonstration illustrates each point developed in the morning lectures. At the end of the afternoon, the saturation threshold of the attendees is generally reached, so the evenings are devoted to discovering Bordeaux wines and vineyards, which helps the otherwise shy attendees enter into discussions concerning femtosecond science.

A document including all the lectures is always distributed to the participants. Step by step this document has been improved as a result of feedback from the attendees and lecturers, who were forced to find pedagogic answers to the many questions arising during the courses. The result is a very comprehensive textbook that we decided to make available to the wider scientific community; i.e., the result is this book.

The people who will gain the most from this book are the scientists (graduate students, engineers, researchers) who are not necessarily trained as laser scientists but who want to use femtosecond pulses and/or gain a real understanding of this tool. Laser specialists will also find the book useful, particularly if they have to teach the subject to graduate or PhD students. For every reader, this book provides a simple progressive and pedagogic approach to this field. It is particularly enhanced by the descriptions of basic experiments or exercises that can be used for further study or practice.

The first chapter simply recalls the basic laser principles necessary to understand the generation process of ultrashort pulses. The second chapter is a brief introduction to the basics behind the experimental problems generated by ultrashort laser pulses when they travel through different optical devices or samples. Chapter 3 describes how ultrashort pulses are generated independently of the laser medium. In Chaps. 4 and 5 the main laser sources used to generate ultrashort laser pulses and their characteristics are described. Chapter 6 presents the different methods currently used to characterize these pulses, and Chap. 7 describes how to change these characteristics (pulse duration, amplification, wavelength tuning, etc.). The rest of the book is devoted to applications, essentially the different experimental methods based on the use of ultrashort laser pulses. Chapter 8 describes the principal spectroscopic methods, presenting some typical results, and Chap. 9 addresses mainly the problems that may arise when the pulse duration is as short as the coherence time of the sample being studied. Chapter 10 describes typical applications of ultrashort laser pulses for the characterisation of electronic devices and the electromagnetic pulses generated at low frequency. Chapter 11 is an overview of the coherent control physical processes making it possible to control evolution channels in atoms, molecules and solids. Several examples of oriented reactions in this chapter illustrate the possible applications of such a technique. Chapter 12 introduces the attosecond pulse generation by femtosecond pulse-matter interaction. It is designed for a best understanding of the physics

principles sustaining attosecond pulse creation as well as the encountered difficulties in such processes.

I would like to acknowledge all persons and companies whose names do not directly appear in this book but whose participation has been essential to the final goal of this adventure. My colleague Gediminas Jonusauskas was greatly involved in the design of the experiments presented during the courses and at the end of the chapters in this book. Danièle Hulin, Jean-René Lalanne and Arnold Migus gave much time during the initial stages, particularly in writing the first version of the course document. The publication of this book would not have been possible without their important support and contribution. My colleagues Eric Freysz, François Dupuy, Frederic Adamietz and Patricia Segonds also participated in the organization of the courses, as did the post-doc and PhD students Anatoli Ivanov, Corinne Rajchenbach, Emmanuel Abraham, Bruno Chassagne and Benoit Lourdelet.

Essential financial support and participation in the courses, particularly by the loan of equipment, came from the following laser or optics companies: B.M. Industries, Coherent France, Hamamatsu France, A.R.P. Photonetics, Spectra-Physics France, Optilas, Continuum France, Princeton Instruments SA and Quantel France.

I hope that every reader will enjoy reading this book. The best result would be if they conclude that femtosecond pulses are wonderful tools for scientific investigation and want to use them and know more.

Bordeaux, April 2004 *Claude Rullière*

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1

Laser Basics

C. Hirlimann

With 18 Figures

1.1 Introduction

Lasers are the basic building block of the technologies for the generation of short light pulses. Only two decades after the laser had been invented, the duration of the shortest produced pulse had shrunk down six orders of magnitude, going from the nanosecond regime to the femtosecond regime. "Light amplification by stimulated emission of radiation" is the misleading meaning of the word "laser". The real instrument is not only an amplifier but also a resonant optical cavity implementing a positive feedback between the emitted light and the amplifying medium. A laser also needs to be fed with energy of some sort.

1.2 Stimulated Emission

Max Planck, in 1900, found a theoretical derivation for the experimentally observed frequency distribution of black-body radiation. In a very simplified view, a black body is the thermal equilibrium between matter and light at a given temperature. For this purpose Planck had to divide the phase space associated with the black body into small, finite volumes. Quanta were born. The distribution law he found can be written as

$$I(\omega)\,\mathrm{d}\omega = \frac{\hbar\omega^3\,\mathrm{d}\omega}{\pi^2c^2(\mathrm{e}^{\hbar\omega/kT}-1)}, \tag{1.1}$$

where $I(\omega)$ stands for the intensity of the angular frequency distribution in the small interval $\mathrm{d}\omega$, $\hbar = h/2\pi$, h is a constant factor which was later named after Planck, k is Boltzmann's constant, T is the equilibrium temperature and c the velocity of light in vacuum. Planck first considered his findings as a heretical mathematical trick giving the right answer; it took him sometime to realize that quantization has a physical meaning.

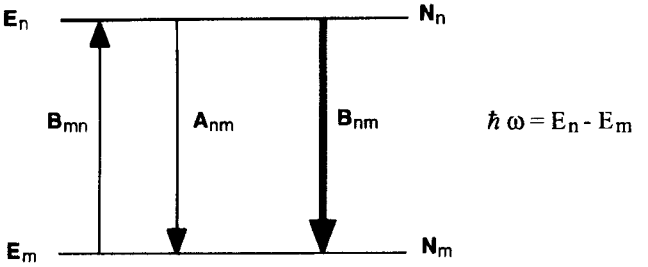


Fig. 1.1. Energy diagram of an atomic two-level system. Energies E_m and E_n are measured with reference to some lowest level

In 1905, Albert Einstein, though, had to postulate the quantization of electromagnetic energy in order to give the first interpretation of the photo-electric effect. This step had him wondering for a long time about the compatibility of this quantization and Planck's black-body theory. Things started to clarify in 1913 when Bohr published his atomic model, in which electrons are constrained to stay on fixed energy levels and may exchange only energy quanta with the outside world. Let us consider (see Figure 1.1) two electronic levels n and m in an atom, with energies E_m and E_n referenced to some fundamental level; one quantum of light, called a photon, with energy $\hbar\omega = E_n - E_m$, is absorbed with a probability B_{mn} and its energy is transferred to an electron jumping from level m to level n. There is a probability A_{nm} that an electron on level n steps down to level m, emitting a photon with the same energy. This spontaneous light emission is analogous to the general spontaneous energy decay found in classical mechanical systems. In the year 1917, ending his thinking on black-body radiation, Einstein came out with the postulate that, for an excited state, there should be another de-excitation channel with probability B_{nm}: the "induced" or "stimulated" emission. This new emission process only occurs when an electromagnetic field $\hbar\omega$ is present in the vicinity of the atom and it is proportional to the intensity of the field. The quantities A_{nm}, B_{nm}, B_{mn} are called Einstein's coefficients.

Let us now consider a set of N atoms, of which N_m are in state m and N_n in state n, and assume that this set is illuminated with a light wave of angular frequency ω such that $\hbar\omega = E_n - E_m$, with intensity $I(\omega)$. At a given temperature T, in a steady-state regime, the number of absorbed photons equals the number of emitted photons (equilibrium situation of a black body). The number of absorbed photons per unit time is proportional to the transition probability B_{mn} for an electron to jump from state m to state n, to the incident intensity $I(\omega)$ and to the number of atoms in the set N_m. A simple inversion of the role played by the indices m and n gives the number of electrons per unit time relaxing from state n to state m by emitting a photon under the influence of the electromagnetic field. The last contribution to the interaction, spontaneous emission, does not depend on the intensity but only

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