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(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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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

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

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.

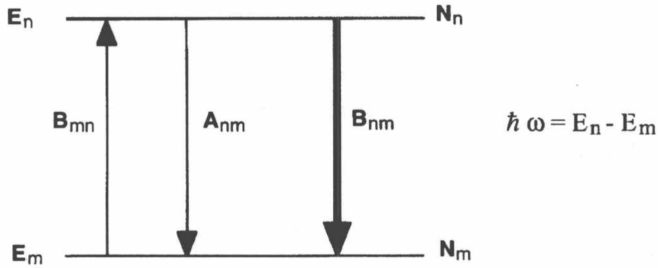

Fig. 1.1. Energy diagram of an atomic two-level system. Energies E_m and E_n are measured with reference to some lowest level

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 on the number of electrons in state n and on the transition probability A_{mn}. This can be simply formalized in a simple energy conservation equation

$$N_m B_{mn} I(\omega) = N_n B_{nm} I(\omega) + N_n A_{nm}. \tag{1.2}$$

Boltzmann's law, deduced from the statistical analysis of gases, gives the relative populations on two levels separated by an energy $\hbar\omega$ at temperature T, $N_n/N_m = \exp(-\hbar\omega/kT)$. When applied to (1.2) one gets

$$B_{mn} I(\omega)\, \mathrm{e}^{\hbar\omega/kT} = A_{nm} + B_{nm} I(\omega) \tag{1.3}$$

and

$$I(\omega) = \frac{A_{nm}}{B_{mn}\, \mathrm{e}^{\hbar\omega/kT} - B_{nm}}. \tag{1.4}$$

This black-body frequency distribution function is exactly equivalent to Planck's distribution (1.1). At this point it is important to notice that Einstein wouldn't have succeeded without introducing the stimulated emission. Comparison of expressions (1.1) and (1.4) shows that $B_{mn} = B_{nm}$: for a photon the probability to be absorbed equals the probability to be emitted by stimulation. These two effects are perfectly symmetrical; they both take place when an electromagnetic field is present around an atom.

Strangely enough, by giving a physical interpretation to Planck's law based on photons interacting with an energy-quantized matter, Einstein has made the spontaneous emission appear mysterious. Why is an excited atom not stable? If light is not the cause of the spontaneous emission, then what is the hidden cause? This point still gives rise to a passionate debate today about the role played by the fluctuations of the field present in the vacuum. Comparison of expressions (1.1) and (1.4) also leads to $A_{nm}/B_{nm} = \hbar\omega^3/\pi^2 c^2$, so that when the light absorption probability is known then the spontaneous and stimulated emission probabilities are also known.

According to Einstein's theory, three different processes can take place during the interaction of light with matter, as described below.

1.2.1 Absorption

In this process one photon from the radiation field disappears and the energy is transferred to an electron as potential energy when it changes state from E_m to E_n. The probability for an electron to undergo the absorption transition is B_{mn}.

1.2.2 Spontaneous Emission

When being in an excited state E_n, an electron in an atom has a probability A_{nm} to spontaneously fall to the lower state E_m. The loss of potential energy gives rise to the simultaneous emission of a photon with energy $\hbar\omega = E_n - E_m$. The direction, phase and polarization of the photon are random quantities.

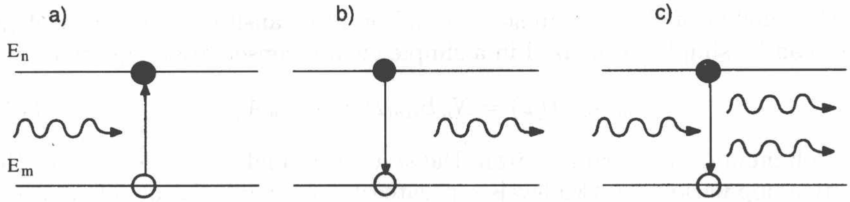

Fig. 1.2. The three elementary electron–photon interaction processes in atoms: (**a**) absorption, (**b**) spontaneous emission, (**c**) stimulated emission

1.2.3 Stimulated Emission

This contribution to light emission only occurs under the influence of an electromagnetic wave. When a photon with energy $\hbar\omega$ passes by an excited atom it may stimulate the emission by this atom of a twin photon, with a probability B_{nm} strictly equal to the absorption probability B_{mn}. The emitted twin photon has the same energy, the same direction of propagation, the same polarization state and its associated wave has the same phase as the original inducing photon. In an elementary stimulated emission process the net optical gain is two.

1.3 Light Amplification by Stimulated Emission

In what follows we will discuss the conditions that have to be fulfilled for the stimulated emission to be used for the amplification of electromagnetic waves.

What we need now is a set of N atoms, which will simulate a two-level material. The levels are called E_1 and E_2 (Fig. 1.3).

Their respective populations are N_1 and N_2 per unit volume; the system is illuminated by a light beam of n photons per second per unit volume with individual energy $\hbar\omega = E_2 - E_1$. The absorption of light in this medium is proportional to the electronic transition probability, to the number of photons at position z in the medium and to the number of available atoms in state 1 per unit volume.

To model the variation of the number of photons n as a function of the distance z inside the medium, the use of energy conservation leads to the

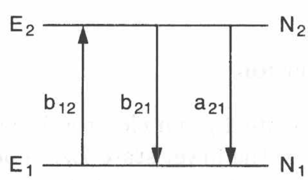

Fig. 1.3. Energy diagram for a set of atoms with two electronic levels

following differential equation:

$$\frac{\mathrm{d}n}{\mathrm{d}z} = (N_2 - N_1)b_{12}n + a_{21}N_2, \tag{1.5}$$

where $b_{12} = b_{21}$ and a_{21} are related to the Einstein coefficients by constant quantities. For the sake of simplicity we will neglect the spontaneous emission process and thus the number of photons as a function of the propagation distance is given as

$$n(z) = n_0\,\mathrm{e}^{(N_2-N_1)b_{12}z}, \tag{1.6}$$

with $n_0 = n(0)$ being the number of photons impinging on the medium.

When $N_2 < N_1$, expression (1.6) simply reduces to the usual Beer–Lambert law for absorption, $n(z) = n_0\,\mathrm{e}^{-\alpha z}$, where $\alpha = (N_1 - N_2)b_{12} > 0$ is the linear absorption coefficient. This limit is found with any absorbing material at room temperature: there are more atoms in the ground state ready to absorb photons than atoms in the excited state able to emit a photon.

When $N_1 = N_2$, expression (1.6) shows that the number of photons remains constant along the propagation distance. In this case the full symmetry between absorption and stimulated emission plays a central role: the elementary absorption and stimulated emission processes are balanced. If spontaneous emission had been kept in expression (1.6), a slow increase of the number of photons with distance would have been found due to the spontaneous creation of photons.

When $N_2 > N_1$, there are more excited atoms than atoms in the ground state. The population is said to be "inverted". Expression (1.6) can be written $n(z) = n_0\,\mathrm{e}^{gz}$, $g = (N_1 - N_2)b_{12}$ being the low-signal gain coefficient. This process is very similar to a chain reaction: in an inverted medium each incoming photon stimulates the emission of a twin photon and its descendants too. The net growth of the number of photons is exponential but does not exactly correspond to the fast doubling every generation mentioned at the end of Sect. 1.2.3. Because the emitted photons are resonant with the two-level system, some of them are reabsorbed; also, some of the electrons available in the excited state are lost for stimulated emission because of their spontaneous decay. The elementary growth factor is therefore less than 2.

1.4 Population Inversion

To build an optical oscillator, the first step is to find how to amplify light waves, and we have just seen that amplification is possible under the condition that there exist some way to create an inverted population in some material medium.

1.4.1 Two-Level System

Let us first consider, again, the two-electronic-level system (Fig. 1.3). Electrons, because they have wave functions that are antisymmetric under inter-