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Excerpt
More information

Part I

Basic concepts

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High-intensity laser–atom physics

In recent years, intense laser fields have become available, over a wide frequency range, in the form of short pulses. Such laser fields are strong enough to compete with the Coulomb forces in controlling the dynamics of atomic systems. As a result, atoms in intense laser fields exhibit new properties that have been discovered via the study of *multiphoton processes*. After some introductory remarks in Section 1.1, we discuss in Section 1.2 how intense laser fields can be obtained by using the "chirped pulse amplification" method. In the remaining sections of this chapter, we give a survey of the new phenomena discovered by studying three important multiphoton processes in atoms: multiphoton ionization, harmonic generation and laser-assisted electron–atom collisions.

1.1 Introduction

If radiation fields of sufficient intensity interact with atoms, processes of higher order than the single-photon absorption or emission play a significant role. These higher-order processes, called multiphoton processes, correspond to the net absorption or emission of more than one photon in an atomic transition. It is interesting to note that, in the first paper he published in *Annalen der Physik* in the year 1905, his "Annus mirabilis," Einstein [1] not only introduced the concept of "energy quantum of light" – named "photon" by Lewis [2] in 1926 – but also mentioned the possibility of multiphoton processes occurring when the intensity of the radiation is high enough, namely "if the number of energy quanta per unit volume simultaneously being transformed is so large that an energy quantum of emitted light can obtain its energy from several incident energy quanta." Multiphoton processes were also considered in the pioneering work of Göppert-Mayer [3].

There are several types of multiphoton processes. For instance, an atom can undergo a transition from a bound state to another bound state of higher energy via the absorption of n photons ($n \geq 2$), a process known as *multiphoton excitation*.

Also, an atom in an excited state can emit n photons in a transition to a state of lower energy, a process called *multiphoton de-excitation*, either by spontaneous emission (which does not require the presence of an external radiation field) or by stimulated emission. Another example is the *multiphoton ionization* (MPI) of an atom, a process in which the atom absorbs n photons, and one or several of its electrons are ejected. An atom interacting with a strong laser field can also emit radiation at higher-order multiples, or harmonics, of the frequency of the laser; this process is known as *harmonic generation*. Finally, radiative collisions involving the exchange (absorption or emission) of n photons can occur in *laser-assisted atomic collisions* such as electron–atom or atom–atom collisions in the presence of a laser field.

Except for spontaneous emission, which will not be considered here, the observation of multiphoton transitions requires relatively large laser intensities. Typically, intensities of the order of $10^8\,\mathrm{W\,cm^{-2}}$ are required to observe multiphoton transitions in laser-assisted electron–atom collisions, while intensities of $10^{10}\,\mathrm{W\,cm^{-2}}$ are the minimum necessary for the observation of multiphoton ionization in atoms. In fact, such intensities are now considered to be rather modest. Indeed, as we shall see in the following section, laser fields have become available in the form of short pulses having intensities of the order of, or exceeding, the atomic unit of intensity

$$I_\mathrm{a} = \frac{1}{2}\epsilon_0 c \mathcal{E}_\mathrm{a}^2 \simeq 3.5 \times 10^{16}\,\mathrm{W\,cm^{-2}}, \tag{1.1}$$

where c is the velocity of light in vacuo, ϵ_0 is the permittivity of free space and $\mathcal{E}_\mathrm{a}$ is the atomic unit of electric field strength, namely

$$\mathcal{E}_\mathrm{a} = \frac{e}{(4\pi\epsilon_0)a_0^2} \simeq 5.1 \times 10^9\,\mathrm{V\,cm^{-1}}, \tag{1.2}$$

where e is the absolute value of the electron charge and a_0 is the first Bohr radius of atomic hydrogen. Atomic units (a.u.) are discussed in the Appendix. We note that $\mathcal{E}_\mathrm{a}$ is the strength of the Coulomb field experienced by an electron in the first Bohr orbit of the hydrogen atom. Laser fields having intensities of the order of, or larger than, I_a are strong enough to compete with the Coulomb forces in governing the dynamics of atoms. Thus, while multiphoton processes involving laser fields with intensities $I \ll I_\mathrm{a}$ can be studied by using perturbation theory, the effects of laser fields with intensities of the order of, or exceeding, I_a must be analyzed by using non-perturbative approaches.

In Chapter 2, we shall discuss the theory of laser–atom interactions based on a semi-classical approach which provides the framework for studying atomic multiphoton processes in intense laser fields. In particular, we shall introduce the *dipole approximation*, in which the laser field is described by a spatially homogeneous electric-field component, while its magnetic-field component vanishes. The dipole approximation is fully adequate to investigate atomic multiphoton processes over a wide range of laser frequencies and intensities. However, as the intensity increases beyond critical values that depend on the frequency, *non-dipole* effects due to the magnetic-field component of the laser field, and eventually *relativistic* effects, must be taken into account [4–7].

The theoretical methods required to solve the quantum-mechanical wave equations introduced in Chapter 2 will be developed in the second part of this book (Chapters 3–7). We shall discuss powerful *ab initio* methods such as the Sturmian-Floquet method [8, 9], the R-matrix–Floquet method [10, 11] and the numerical solution of the time-dependent Schrödinger equation [12, 13]. In this second part, we shall also examine *methods of approximation* which can be used to analyze multiphoton processes at low or at high laser frequencies, respectively. All of these methods will be applied in the third part (Chapters 8–10) to analyze atomic multiphoton processes.

The subject of atoms in intense laser fields has been covered in the volumes edited by Gavrila [14] and by Brabec [15], in the review articles by Burnett, Reed and Knight [16], Joachain [17], Kulander and Lewenstein [18], Protopapas, Keitel and Knight [19], Joachain, Dörr and Kylstra [20], Milosevic and Ehloztky [21], and also in the books by Faisal [22], Mittleman [23], Delone and Krainov [24] and Grossmann [25].

1.2 High-intensity lasers

To obtain high-intensity laser fields, one must concentrate large amounts of energy into short periods of time, and then focus the laser light onto small areas. In an intense laser system, the oscillator produces a train of pulses of short duration. The amplifier then increases the energy of the pulses, which are subsequently focused. A very successful method of amplification, called "chirped pulse amplification" (CPA), was devised in 1985 by Strickland and Mourou [26]. This method, which is illustrated in Fig. 1.1, consists in the following three steps. Firstly, the short laser pulse to be amplified (produced by the oscillator) is stretched in time into its frequency components by a dispersive system such as a pair of diffraction gratings, so that a *chirped* pulse is generated. This stretching in time of the pulse greatly reduces its peak intensity, so that in the second step the frequency components of the chirped pulse can be sent in succession through a laser amplifier without distortions and damage. In the third step, the amplified chirped pulse is compressed in time by another pair of diffraction gratings, which recombine the dispersed frequencies, thus producing a short pulse with a very large peak intensity. Finally, the resulting amplified short pulse is tightly focused onto a small area. After focusing, intensities of the order of the atomic unit of intensity I_a can be readily obtained. An important advantage of the CPA method is that it can yield very intense, short pulses by using

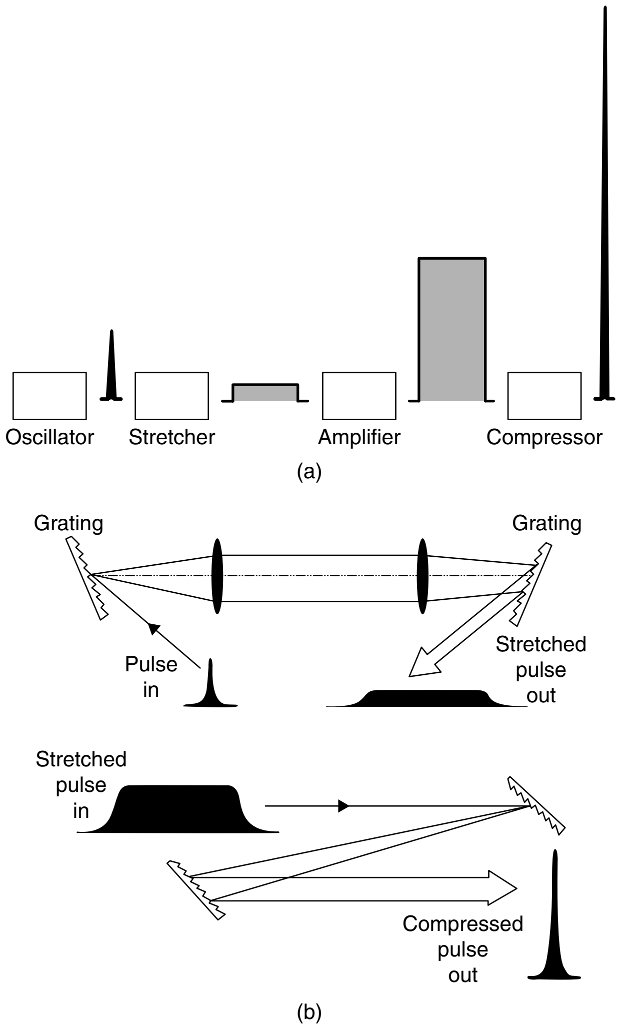

Figure 1.1. Chirped pulse amplification (CPA) method. (a) An oscillator produces a short pulse, which is then chirped (stretched in time into its frequency components). In this way, the peak intensity of the pulse is lowered, so that amplification can take place without damage or distortions. The amplified chirped pulse is then compressed in time, resulting in a short pulse with a very high intensity. (b) The matched stretcher and compressor of the CPA method. The stretcher (top) consists of a telescope of magnification unity placed between two antiparallel gratings. In this configuration, the low-frequency components of the pulse have a shorter optical path than the high-frequency ones. Conversely, the compressor (bottom) consists of a pair of parallel gratings, so that the optical path for the high-frequency components of the pulse is shorter than for the low-frequency ones. (From G. A. Mourou, C. P. J. Barty and M. D. Perry, *Phys. Today*, **Jan.**, 22 (1998).)

a "table-top" laser system. A review of the CPA method has been given by Mourou, Tajima and Bulanov [27].

The first CPA high-intensity lasers to be constructed used Nd:glass as the amplifying medium. In the system developed during the 1990s at Imperial College, London, a pulse from a Nd oscillator, of wavelength $\lambda = 1064$ nm, duration 1 ps (10^{-12} s) and energy 1 nJ was stretched by diffraction gratings to about 25 ps. It was then amplified by using Nd:glass as the amplifying medium to an energy of about 1 J. This amplified chirped pulse was subsequently compressed to a duration close to its initial picosecond value by diffraction gratings, so that output powers of around 1 TW (10^{12} W) could be obtained. By focusing over an area having a diameter of 10 μm, intensities of the order of 10^{18} W cm^{-2} were reached. Lasers of this kind have a repetition rate of about one shot per minute. More recently, CPA laser systems employing Ti:sapphire for the oscillator and the amplifying medium have been used extensively, because they can generate very short pulses with high repetition rates. If only moderate intensities ($\sim 10^{14}$ W cm^{-2}) are required, such laser systems can produce pulses having a duration of about 30 fs, with a repetition rate of 300 kHz. CPA Ti:sapphire lasers can also yield very intense pulses. For example, at the ATLAS laser facility in the Max-Planck Institut für Quantenoptik in Garching, pulses of 100 fs in duration and 1 nJ in energy have been stretched, amplified and compressed, giving output pulses of wavelength $\lambda = 790$ nm, duration 150 fs and energy 220 mJ at a repetition rate of 10 Hz. After focusing on a spot 6 μm in diameter, the intensity available from this laser reached 4×10^{18} W cm^{-2}. More recently, intensities up to 10^{22} W cm^{-2} have been achieved using the Hercules Ti:sapphire laser at the University of Michigan [28].

The CPA concept was originally developed for the amplification of short laser pulses with laser amplifiers based on laser gain media. However, it was subsequently realized that it can also be used with optical parametric amplifiers (OPA), in which case it is known as the *optical parametric chirped pulse amplification* (OPCPA) method [29, 30]. Optical parametric amplification [31, 32] is a second-order phenomenon of non-linear optics, arising from the fact that crystal materials lacking inversion symmetry can display a $\chi^{(2)}$ non-linearity, where $\chi^{(n)}$ denotes the nth-order susceptibility [33]. Apart from other effects (frequency doubling, generation of sum and difference frequencies), this gives rise to parametric amplification, in which a weak signal beam of angular frequency ω_1 and an intense pump beam of angular frequency $\omega_3 > \omega_1$ generate two intense beams with angular frequencies ω_1 and $\omega_2 = \omega_3 - \omega_1$. Indeed, as the signal beam and the pump beam propagate together through the crystal, photons of the pump beam, having energy $\hbar\omega_3$, are converted into lower-energy signal photons of energy $\hbar\omega_1$ and an equal number of "idler" photons of energy $\hbar\omega_2 = \hbar(\omega_3 - \omega_1)$, where $\hbar = h/(2\pi)$ and h is Planck's constant. A schematic diagram of an optical parametric amplifier is shown in Fig. 1.2.

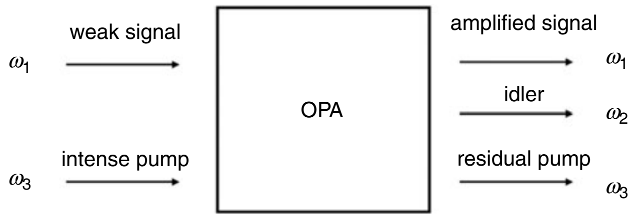

Figure 1.2. Optical parametric amplifier.

Recent technological advances in ultra-fast optics have allowed the generation of intense laser pulses comprising only a few optical cycles (that is, laser periods, where the laser period is defined as $T = 2\pi/\omega$) of the laser field [34, 35]. In particular, the development of Ti:sapphire laser systems using the CPA method has made possible the generation of such high-intensity "few-cycle pulses" in the near infra-red region of the electromagnetic spectrum, with a central wavelength around 800 nm, corresponding to a photon energy of 1.55 eV and an optical cycle of 2.7 fs.

A successful way of obtaining intense few-cycle laser pulses relies on external bandwidth broadening of amplified pulses in gas-filled capillaries [36, 37], using the hollow-fiber technique [38] and chirped-mirror technology [39]. For example, using a Ti:sapphire laser system and a hollow-fiber chirped-mirror compressor, Sartania *et al.* [37] demonstrated the generation of 0.1 TW, 5 fs laser pulses at a repetition rate of 1 kHz. However, the method of gas-filled capillaries involves important energy losses and is difficult to scale to very high energies and peak powers. Several OPCPA systems delivering very intense, few-cycle 800 nm laser pulses have also been reported [40–43].

In addition to very high peak intensities and high repetition rates, the laser systems delivering few-cycle pulses must also provide reliable control over the *carrier-envelope phase* (CEP) φ, namely the phase of the carrier wave with respect to the maximum of the laser pulse envelope, since the CEP sensitively determines the variation of the electric field [44].

As an example, we show in Fig. 1.3 the wave form of the electric field of a linearly polarized laser pulse whose carrier wavelength is $\lambda = 800$ nm and whose intensity profile is proportional to $F(t)\cos(\omega t + \varphi)$, where $F(t)$ is a sech envelope function. In Fig. 1.3(a), the CEP is $\varphi = 0$, corresponding to a "cosine-like" pulse, while in Fig. 1.3(b) the CEP is $\varphi = -\pi/2$, corresponding to a "sine-like" pulse.

Intense few-cycle laser pulses with stabilized CEP have been obtained by using CPA Ti:sapphire laser systems [45–56]. A few-cycle OPCPA system producing infra-red laser pulses at a wavelength of 2.1 μm with a stable CEP has also been demonstrated [57]. One of the major goals is to perform a *single-shot* determination

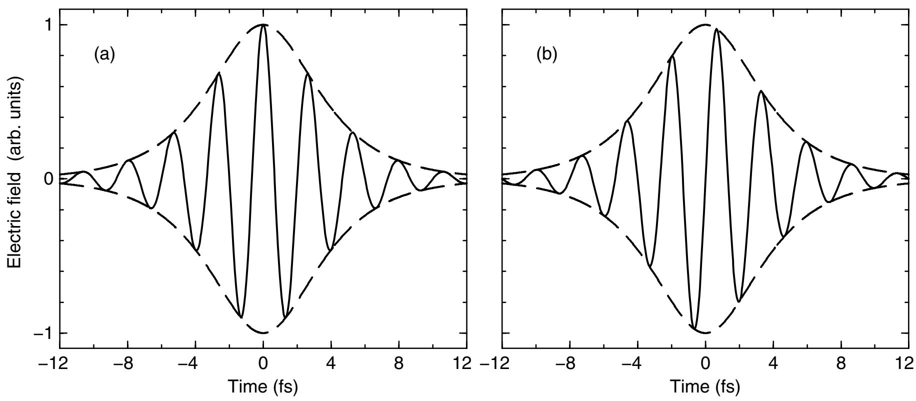

Figure 1.3. Wave form of the electric field (solid curves) of a linearly polarized laser pulse, taken to be proportional to $F(t)\cos(\omega t + \varphi)$, where $F(t)$ is a sech envelope function such that $F^2(t)$ is a sech^2 function of 5 fs full width at half maximum (dashed curves). The carrier wavelength is $\lambda = 800$ nm and the carrier-envelope phase is (a) $\varphi = 0$ for the "cosine-like" pulse and (b) $\varphi = -\pi/2$ for the "sine-like" pulse.

of the CEP of the laser system, while using only a relatively small fraction of the available laser pulse energy.

Intense few-cycle laser pulses with a stable CEP play an important role in high-intensity laser–matter interactions. Indeed, with such pulses, complete control of the electric field wave form of the laser pulse is obtained, since the pulse shape, the carrier wavelength and the carrier-envelope phase can all be determined. As a result, these pulses provide a new way to study the electron dynamics in intense laser–atom processes. They can exert a controlled force on electrons that may vary on atomic scales, not only in strength, but also in time.

Most of the work in the area of high-intensity laser–matter interactions has been restricted to infra-red, visible and ultra-violet radiation [58, 59]. With the advent of *free electron lasers*, another source has become available to perform experiments over a wide range of wavelengths extending from the millimeter to the X-ray domains [60].

In a free-electron laser (FEL), an electron beam moving at a relativistic velocity passes through a periodic, transverse magnetic field produced by arranging magnets with alternating poles along the beam path. This array of magnets is called an undulator or wiggler because it forces the electrons to acquire a wiggle motion in the plane orthogonal to the magnetic field. This transverse acceleration produces spontaneous longitudinal emission of electromagnetic radiation of the synchrotron radiation type. Laser action is due to the fact that the electron motion is in phase with the electromagnetic field of the radiation already emitted, so that the fields add coherently and further emission is stimulated. Free-electron lasers have many attractive properties such as wide tunability and high laser power. However, they are large and expensive, since they involve using electron beam accelerators. The first FEL was demonstrated at a wavelength of 3.4 μm using the Stanford Linear Accelerator [61]. Since then, several FELs have been operated at wavelengths ranging from the millimeter to the soft X-ray region.

In the short-wavelength (VUV and X-ray) region, the lack of appropriate mirrors prevents the operation of an FEL oscillator. As a result, there must be suitable amplification during a single pass of the electron beam through the undulator. It is worth noting that even if the initial electromagnetic field is zero, laser action can still occur in the FEL through the process of "self-amplified spontaneous emission" (SASE), whereby shot noise in the electron beam causes a noisy signal to be initially radiated. This noise then acts as a seed for the FEL, so that the amplification process develops and intense coherent radiation is produced in a narrow band around the resonance wavelength. The first observation of the SASE process was reported at the Free Electron Laser in Hamburg (FLASH) at a wavelength of 109 nm [62]. Also at the FLASH facility, short VUV laser pulses of wavelengths in the range 95–105 nm, durations of 30–100 fs and peak powers at the gigawatt level have been generated [63]. More recently, lasing was observed by the FLASH team at a wavelength of 6.5 nm, in the soft X-ray domain. The European X-ray Free Electron Laser (XFEL) in Hamburg and the Linac Coherent Light Source (LCLS) at Stanford, both under development, will operate at wavelengths down to around 0.1 nm, well into the X-ray region.

1.3 Multiphoton ionization and above-threshold ionization

In this section, we give a survey of the basic features of the *multiphoton ionization* (MPI) process, starting with the multiphoton *single* ionization reaction

$$n\hbar\omega + A^q \to A^{q+1} + \mathrm{e}^- , \tag{1.3}$$

where q is the charge of the target atomic system A, expressed in atomic units, $\hbar\omega$ is the photon energy and n is a positive integer.

This process was first observed in 1963 by Damon and Tomlinson [64], who used a ruby laser to ionize helium, argon and a neutral air mixture. In subsequent investigations, Voronov and Delone [65] used a ruby laser to induce seven-photon ionization of xenon, and Hall, Robinson and Branscomb [66] recorded two-photon electron detachment from the negative ion I^-. In later years, important results were obtained by several experimental groups, in particular at Saclay, where the dependence of the ionization rates on the laser intensity were studied. For the intensities $I \ll I_\mathrm{a}$ available at that time, it was observed that the total n-photon