# 10: Atoms in Strong Fields

**Objectives**

In this chapter the excitation of atoms by intense laser pulses is discussed. This topic has grown to be one of the most exciting fields of research in atomic, molecular and optical (AMO) physics. Nowadays high-power lasers are accessible to many laboratories giving the possibility to investigate the interesting nonlinear effects which may appear.

**Prerequisites**

The structure of atoms. Electromagnetic field intensity, anharmonic oscillations.

## Introduction

The field of research "atoms in strong laser fields" was born a few years after the invention of the laser in 1960, and has evolved considerably during the last three decades owing to the rapid technological development of high-power short-pulse lasers. Early experiments had shown that the response of an atomic system started to be nonlinear at around $I_n = 10^8 \mathrm{W/cm^2}$ for visible or near-infrared laser fields, which could be characterized by second, third or fourth order nonlinear susceptibilities. Theoretically this range can be described by the appropriate quantum mechanical method, called perturbation theory. The next important limiting intensity is $I_s = 10^{13} \mathrm{W/cm^2}$, above which the traditional quantum mechanical approaches of treating nonlinear optical processes already fails, and the interaction becomes strongly nonlinear, necessitating new concepts.

An even higher and rather obvious new limit is, when a laser field is strong enough to overcome the Coulomb forces in the atom. In order to see this, consider the field of the proton at the distance of the first Bohr radius $a0$, which is:

$$\mathcal{E}_a = \frac{|q_0|}{4\pi\epsilon_0}\frac{1}{a_0^2} = 5.1 \times 10^9 \ \mathrm{V/cm} \quad (10.1)$$

The intensity of a laser field where the oscillating electric field amplitude has the same value is:

$$I_a = \frac{1}{2}\epsilon_0 c \mathcal{E}_a^2 = 3.5 \times 10^{16} \ \mathrm{W/cm^2} \quad (10.2)$$

**Problem 10.1**

The intensity can also be given by the photon flux, which is:

$$\phi = \frac{I}{\hbar\omega} \quad \mathrm{photon/(s \cdot cm^2)} \quad (10.3)$$

Calculate the photon flux of a $800\mathrm{nm}$ Ti-Sapphire laser for $I_a$.

## Experiments with intense pulses

As the most powerful laser material namely titanium-sapphire, and a new amplification technique, chirped pulse amplification, have become common around 1990, it made high-power lasers accessible to many laboratories. Since then this topic has grown to be one of the most exciting fields of research in atomic, molecular and optical (AMO) physics. A typical and general experimental setup for studying atoms in strong laser fields is shown in Figure 10.1.

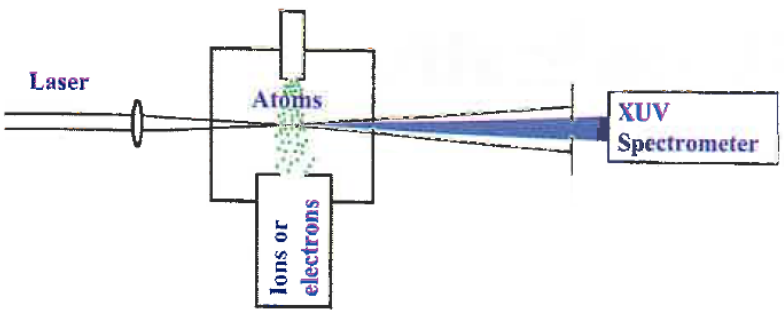



Figure 10.1: Typical schematic experimental setup in the study of atoms in strong laser fields. An intense short pulse laser is focused into an interaction chamber, which contains a gas of atoms. Ions or electrons can be detected, e.g. with time-of-flight techniques.

The first element is a short pulse laser. The laser pulses used are often in the femtosecond range, the shortest being sub-10fs. High repetition rates, today in the kHz range, allow experimentalists to get good statistics. Focused intensities needed to get into the strong field regime are of the order of $10^{14}-10^{15}\mathrm{W/cm^2}$. Since about 1995 the favourite tool has become the titanium sapphire laser, operating at $800\mathrm{nm}$ in the near-infrared, and providing very short pulse duration, high laser intensity at high repetition rate. The second part of the experimental setup is a vacuum chamber where a gas of atoms (often rare gases) is being introduced, in a cell or pulsed jet. One measures the energy of the ions generated or that of the emitted photoelectrons. In addition, angular distribution of the photoelectrons can be recorded. In high-order harmonic generation experiments, the atomic density is much higher than in ionisation experiments, pressures up to a few hundreds mbar is the usual value. The radiation emitted on axis can be detected and analysed using a standard XUV (extreme ultraviolet) spectrometer, including a grating and a photon detector.

In order to classify the strong field effects we first introduce the important quantities that can be used to describe the processes. One of them is the ionisation energy or (ionisation potential) denoted by $I_P$, and shown in Figure 10.2

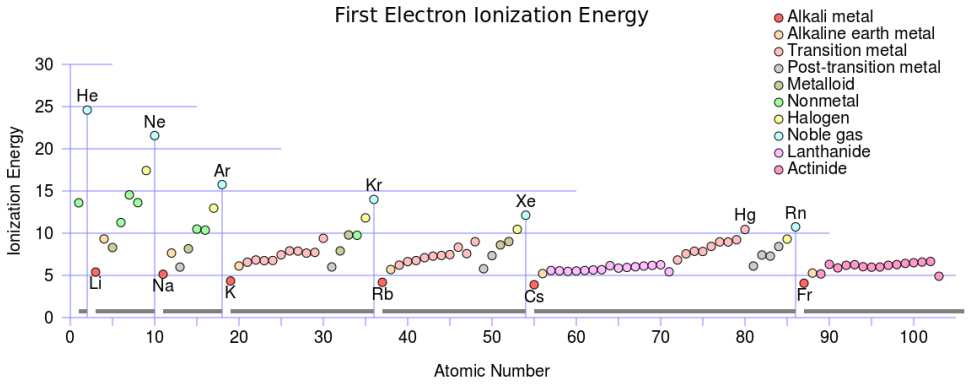



**Figure 10.2**: Ionization energies $I_p$ of atoms, given here as the function of *Z*. It has the highest values for noble gas atoms, as these are the most stable chemical elements, while it is relatively small for alkali metal atoms, which have only a single electron outside a closed electron shell. The value of $I_p$ for the H atom is $13.6\mathrm{eV}$, which is the negative of the ground state energy.
https://en.Wikipedia.org/wiki/File:First_Ionization_Energy.svg

We also introduce here the important notion of the ponderomotive potential $U_P$ of the exciting field. It is the energy gained by an otherwise free electron in a harmonically oscillating field. A simple classical calculation shows that according to the equation of motion

$$m\dot{v} = q_0 E_0 \sin\omega t \quad (10.4)$$

the kinetic energy averaged over a period $2\pi/\omega$ is

$$\overline{\tfrac{1}{2}mv^2} = \tfrac{1}{2}\frac{q_0^2E_0^2}{m\omega^2}\overline{\cos^2\omega t} = \frac{q_0^2E_0^2}{4m\omega^2} =: U_P \quad (10.5)$$

The characteristics of the response of the atom depends on the ratio of the ionisation potential $I_P$ and the ponderomotive potential through the parameter:

$$\gamma_K := \sqrt{\frac{I_P}{2U_P}} \quad (10.6)$$

introduced by L.V. Keldysh in 1964, and called since then the Keldysh parameter.

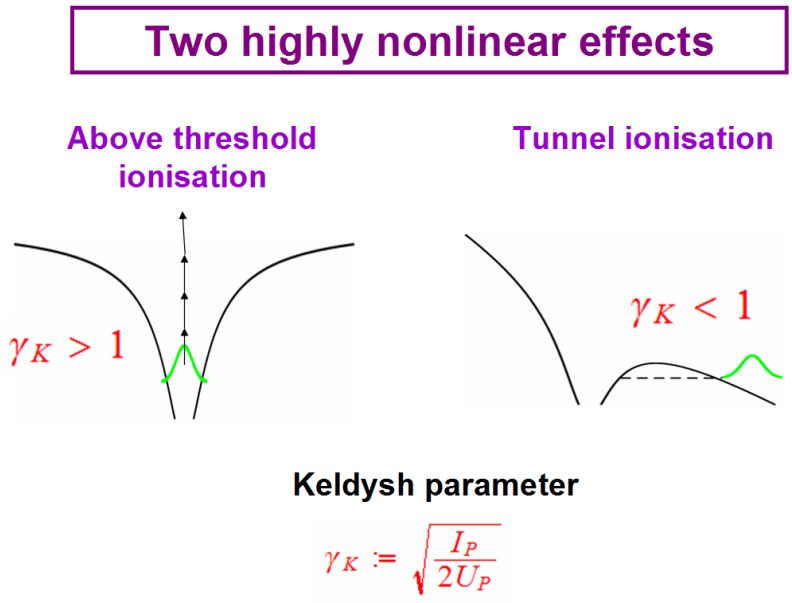



Figure 10.3: The response of the atom depends on the ratio of the ionisation potential $I_P$ and the ponderomotive potential through the Keldysh parameter.

If the external field strength i.e. the square root of the incoming intensity is high enough then $\gamma_K < 1$ and the atomic potential can be significantly changed by that field as shown in the right hand side of the figure 10.3. The electron may then tunnel out through the potential barrier which is now the sum of the atomic and external fields. The barrier can even become lower than the energy of the bound state, then we can speak of **over the barrier ionisation**. This is valid of course for a half period of the oscillating field since the external field changes sign in the next half period and the force on the electron will change its direction to the opposite. For pulses with $\gamma_K > 1$, one observes a different process called above threshold ionisation, shown in the left hand side of the figure, and to be discussed in the next section.

## Above threshold ionisation

Electrons initially in the ground state can absorb a large number of photons, many more than the minimum number required for ionisation, thus being ionised with a high kinetic energy. This new physical effect can be observed at intensities above (I_{s}=10^{13}\mathrm{W}/\mathrm{cm}^{2}\). When energy resolved photoelectron measurements have become possible in the end of the 1970s, it was observed that in the photoionization process the ejected electrons could absorb photons in excess of the minimum number required for ionisation to occur. The photoelectron spectra were seen to exhibit several peaks, separated by the photon energy $\hbar\omega$. The electron energies appear at the values

$$E_s = (n+s)h\omega - I_p \quad (10.7)$$

where $n$ is the minimum number of photons to exceed the ionisation potential $I_P$, and $s = 0, 1, \ldots$ where ss is the number of excess photons, or above threshold photons, absorbed by the electron. This process was shown for the first time in 1979 and it was called **above threshold ionization** (ATI). A typical ATI spectrum shows a number of electron peaks separated by the laser photon energy.

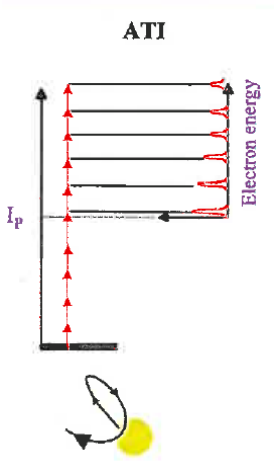



Figure 10.4:

For short and intense laser pulses, the ionisation potential $I_p$ is increased by the (time-dependent) ponderomotive potential, i.e. the mean kinetic energy acquired by an electron oscillating in the laser field $U_P$. The experimental precision in detecting electron spectra increased significantly from the mid 90's owing essentially to higher laser repetition rates. Thus ATI spectra with many decades in number of counts could be recorded. Amazingly, the spectra were found to extend over many tens of eV, with a decrease for the first orders, up to $2U_p$, followed by a large plateau extending to $10U_p$. In general, with linear polarisation, electrons are generated along the polarisation's direction. It was found that angular distributions exhibit a much more complex (off-axis) structure at the edge of the plateau, called "scattering rings". The large plateau and the complex angular structure originate from the rescattering of the electron wavepacket on the parent ion.

### Multiple ionisation

Not only one, but many electrons can be emitted from atoms subject to strong laser fields. Double ionisation of alkaline earth atoms was observed as early as in 1975 and the first evidence for non-sequential ionisation of rare gas atoms was first demonstrated in 1983. They can be emitted one at a time, in a sequential process, or simultaneously, a mechanism called direct, or non-sequential. The simplest multiple ionisation mechanism for atoms in strong laser fields is the so-called sequential stripping mechanism, i.e. a sequence of single electron ionisation acts: ionisation of the atom, then of the singly charged ion, then of the doubly charged ion and so on. The main experimental effort during the 1980's was to test the limits of this mechanism with the available laser powers and to understand the process responsible for the ionisation of the different charge states (multiphoton or tunnelling). It turned out however, that sequential ionisation is not the only mechanism responsible for multiple ionisation. Progress in experimental techniques with, for example, recoil-ion momentum spectroscopy and electron-ion coincidence measurements allows now scientists to record the energies and angular distributions of the electrons emitted during a multiple ionisation process, thus providing better experimental insight.

### High-order harmonic generation

Another effect also observed at sufficiently high intensities was high harmonic generation (HHG). Atoms interacting with a strong laser field can emit radiation at frequencies that are high order multiples of the angular frequency of the pump laser. A high-order harmonic spectrum consists of a sequence of peaks at circular frequencies which are odd multiples of the driving, or fundamental circular frequency:

(10.8)

Only odd orders can be observed, owing to inversion symmetry in an atomic gas. In the time domain, this means that the process is periodic with a periodicity twice the laser period. A HHG spectrum has a characteristic behaviour: it shows a fast decrease for the first few harmonics, followed by a long plateau of harmonics with approximately constant intensity. The plateau ends up by a sharp cut-off. Most of the early work on harmonic generation concentrated on the extension of the plateau, i.e. the generation of harmonics of shorter wavelength. Today, harmonic spectra produced with short and intense laser pulses extend to more than 500 eV, down to the water window below the carbon K-edge at 4.4 nm. The mechanism of the generation is shown in figure 10.5.

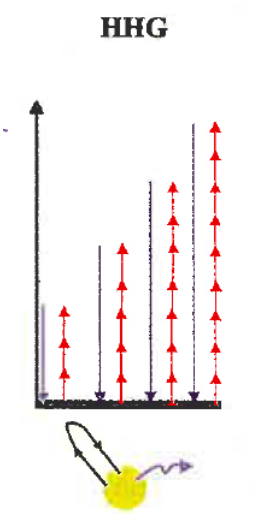



Figure 10.5:

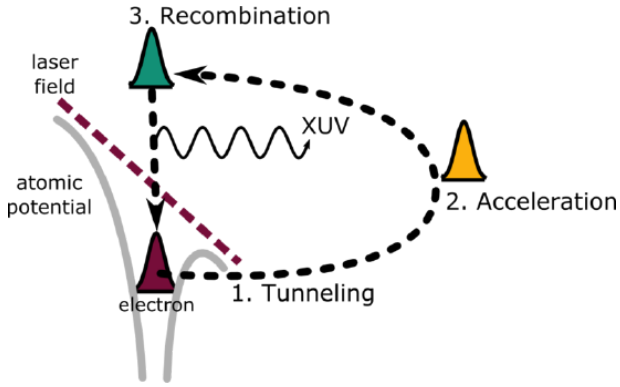



**Figure 10.6**: Simple illustration of the semi-classical three-step model of high-order harmonic generation. http://en.Wikipedia.org/wiki/File:ThreeStep.png

A large effort has been devoted to optimize and characterize the properties of this new source of XUV radiation. A milestone in the understanding of HHG processes was the finding by Kulander and co-workers in 1992 that the cut-off position in the harmonic spectrum follows the universal law $I_p + 3U_p$. This result was interpreted in terms of a simplified theory, called a three step model shown in figure 10.6. In the first step a tunnelling process takes place. In the second step the model neglects the Coulomb force of the core, and assumes that the electron moves only under the effect of the laser field, which first pulls away the electron from the core, but when during its oscillation it changes its sign the field may return the electron back to the core. Thirdly: when slowing down, the electron radiates in the form of harmonics whose energy comes from the energy lost by the returning electron. A realistic description of HHG must involve the calculation of the single atom response, and also the solution of propagation (Maxwell) equations for the emitted radiation.

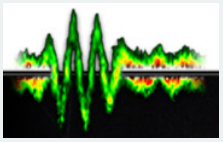
An interesting consequence of the existence of the harmonics is that among favourable circumstances they can add up and produce a sequence of peaks of pulses in the attosecond range (duration ~200 as), to be considered in the next section.

## Attosecond Pulses

Almost immediately after the first observation of the harmonic plateau at the end of the 1980's, it was realised first by Győző Farkas and Csaba Tóth that, if the harmonics were emitted in phase, i.e. phase-locked, then the temporal structure of the radiation emitted from the medium would consist of a "train" of attosecond pulses separated by half the laser period. There is a clear analogy here with mode-locked lasers, where axial modes oscillating in a laser cavity are locked in phase, leading to the production of trains of short pulses. From a microscopic point of view, at each half-laser cycle, there is a short (attosecond) burst of light, as an electron recombines back to the ground state. Isolated attosecond pulses can be produced if one limits these returns to single events. The simplest idea is to use a very short (7 fs) and intense laser pulse. Such a laser source should allow one to generate single attosecond pulses, because harmonic generation occurs during a limited time interval before the onset of ionisation. Attosecond pulses have

remained, however, essentially a theoretical prediction, until 2002. Then an important experiment by the group of Agostini and Muller have shown evidence for trains of 250as pulses, while Ferenc Krausz and coworkers were able to generate isolated 650as650as pulses.

Further Reading



If you are interested in this topic check out the home page of Ferenc Krausz's group. Under the menu "Light and Matter" a lot of interesting material can be found on attosecond physics, light matter interaction etc..
http://www.attoworld.de/Mainpages/Light_and_matter/index.html

## Outlook

The harmonic radiation, with attosecond pulse duration, high brightness and good spatial and temporal coherence, could be used in a growing number of applications ranging from atomic and molecular spectroscopy to solid-state and plasma physics. It has also been proposed as an alternative source for nanolithography, in particular for metrological purposes. It opens up two new fields of research: multiphoton processes in the XUV range, and attosecond physics, where processes in atoms and molecules can be studied at an unprecedented time scale. Attophysics is just born, and there is already an active discussion on the possible applications of attosecond XUV pulses. To produce routinely pulses of duration in the attosecond range is one of the main targets of the Extreme Light Infrastructure Attosecond Light Pulse Source (ELI ALPS) facility to be built up in Szeged, Hungary within the next few years.