

# Ultrashort Laser Pulses Applications

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## 1. Introduction

Ultrashort laser pulses are considered to be pulses of electromagnetic radiation whose duration is shorter than the thermal vibration period of molecules, around tens of picoseconds ( $10^{-12}$  s). Pulses with durations of a few picoseconds were already produced in the 1960's (DiDomenico et al., 1966), shortly after the laser invention, using the mode-locking technique (Hargrove et al., 1964; Haus, 2000). In the next decade, refinements on this pulse generation scheme, and the use of bulky dye lasers with large emission bandwidths, shortened the pulses to the hundreds of femtoseconds ( $10^{-15}$  s) timescale (Diels et al., 1978; Shank & Ippen, 1974). In the 1980's, pulses with durations below 10 femtoseconds were generated from dye lasers (Fork et al., 1987; Knox et al., 1985), however the applications had to wait for the Ti:Sapphire Kerr-Lens mode-locked laser (Brabec et al., 1992; Spence et al., 1991) and the Chirped Pulse Amplification (CPA) technique (Strickland & Mourou, 1985) to really spread out. The large Kerr effect and broad emission bandwidth available in the Ti:Sapphire (Moulton, 1986), and the diode pumped solid state lasers (Keller, 1994; Keller, 2010; Scheps, 2002) that became available around this time, greatly simplified the setup needed to generate ultrashort pulses, and promptly replaced the dye lasers for this purpose. Finally, the invention of the CPA technique in 1985, allowed the generation of high intensity ultrashort pulses in all-solid state laser systems, and disseminated these laser systems due to its simplicity of operation when compared to the preceding systems, stability and relatively low cost. The CPA technique (Diels & Rudolph, 2006; Maine et al., 1988; Mourou et al., 1998) consists in generating ultrashort pulses with nanojoules of energy in a main oscillator laser, then temporally stretching these pulses by chirping its frequency (dispersion control) (Rullière, 1998) to decrease its power and intensity, allowing its amplification up to more than a million times without damaging the amplification chain components; after the amplification, the pulses are compressed to durations close to their original ones and directed to the applications. When the CPA technique became prominent, the pulses stretching and compression were mainly done with diffraction gratings (Fork et al., 1984; Martinez, 1987a; Martinez, 1987b; Treacy, 1969), although nowadays other techniques are disseminated, such as the use of Chirped Mirrors (Nisoli et al., 1997; Szipocs et al., 1994), fiber stretchers (Zhou et al., 2005) and prisms and grisms compressors (Chauhan et al., 2010).

As the years went by, scientific and technological developments led to the dissemination of ultrashort pulses systems based in other mode-locking schemes such as SESAMs (Semiconductor Saturable Absorber Mirrors) (Keller, 2010) and gain media including

chromium, ytterbium and neodymium doped crystals (Diels & Rudolph, 2006), neodymium doped glasses (Badziak et al., 1997), erbium doped fibers (Krauss et al., 2010), Optical Parametric CPAs (OPCPA) (Dubietis et al., 2006) and picosecond semiconductor lasers (Koda et al., 2010). The availability of systems with varying characteristics in many universities and research laboratories resulted in numerous ultrashort laser pulses applications in many areas. The great variety of ultrashort pulses laser systems available nowadays, both commercially and under development in laboratories around the world, is capable of generating pulses with durations in the range from a few femtoseconds to hundreds of femtoseconds, and picoseconds for the semiconductor lasers, wavelengths ranging from the ultraviolet spectrum (Reiter et al., 2010) up to 1.5  $\mu\text{m}$ , energies extending from nanojoules to thousands of Joules (Stoeckl et al., 2006), terawatt peak powers in repetition rates up to few kilohertz (Bagnoud & Salin, 2000), and reaching tens of MHz at lower powers (Malinowski et al., 2004; Naumov et al., 2005). Powers over one petawatt ( $10^{15}$  W) are generated in national laboratories (Chambaret et al., 2007; Gaul et al., 2010; Habara et al., 2010; Musgrave et al., 2007; Perry & Mourou, 1994; Tajima & Mourou, 2002), and intensities up to  $10^{22}$  W/cm<sup>2</sup> (Bahk et al., 2005) were obtained.

Laser oscillators cannot generate pulses shorter than a few femtoseconds due to the wavelengths and bandwidths necessary for this, in the ultraviolet spectrum. Nevertheless, the use of High Harmonic Generation techniques allow the creation of pulses down to a few hundreds of attoseconds ( $10^{-18}$  s) (Sansone et al., 2006).

The applications of ultrashort laser pulses derive mainly from two characteristics of such pulses: firstly, their very short duration, which can be used to induce and measure ultrafast phenomena with temporal resolution of femtoseconds (and more recently with hundreds of attoseconds), besides minimizing heat transfer to the target. Secondly, the huge electric and magnetic fields associated to the pulses that can surpass the ones that bind electrons to atoms, resulting in large ionizations that lead to material modifications; the free electrons generated can also be accelerated by the electromagnetic field to high energies, even into the relativistic regime.

The following sections will explore ultrashort laser pulses applications, roughly going from lower to higher energy (power, intensity) ones. Not all applications will be covered due to space limitations, and references will be given for those wanting a deeper understanding on the various subjects.

## 2. Time resolved measurements

To measure an event, a faster phenomenon is needed, and the extremely short duration of ultrashort pulses make them ideal for measuring processes that occur in the picosecond and femtosecond time scales. Moreover, ultrashort pulses can be used to induce a series of fast phenomena in various kinds of samples with the advantage of shortening the transient behavior resulting from the optical excitation, so the consequences of this perturbation are free to develop in time. The shorter the pulse, the faster the transient behavior vanishes and the excitation evolution can be measured. Ultrashort pulses also have the benefit of minimal heat transfer to the sample, not exciting vibrational modes that can introduce noise or even mask the signal under measurement. The sample can be a dielectric material, whose absorption coefficient is changed due to saturation of this absorption, a semiconductor in which the charge carriers created in the conduction band by the ultrashort pulse change the material properties (Yariv & Yeh, 2007), a metal whose magnetic characteristics are altered

by the ultrashort pulse (Beaurepaire et al., 1996), or even an atom that has its electronic properties modified by the light absorption (Drescher et al., 2002). Electronic process in biological molecules can also be stimulated (Li et al., 2010; Schreier et al., 2007).

When a pulse can be used to induce a phenomenon and to measure its temporal evolution, a pump-probe technique should be used. In this technique (Diels & Rudolph, 2006; Rullière, 1998) two replicas of an ultrashort pulse are created using a beam splitter, one with most of the original pulse energy (pump pulse), and the other one with a only small fraction of that energy (probe pulse), as represented on Fig. 1. Both pulses are focused upon the sample under study overlapping spatially at the focus, and one of the pulses (usually the pump) goes first through a delay line, in which a relative delay,  $\Delta t$ , can be adjusted between the pump and the probe pulses. At  $t = 0$  the pump pulse excites the sample, and at the time  $t = \Delta t$  the weaker pulse goes through the sample and has one or few of its characteristics (intensity, polarization, phase, temporal duration) modified, probing the excitation. Repeating the measurement varying the delay ( $\Delta t$ ) from negative to positive values, determines the temporal dynamics of the excitation.

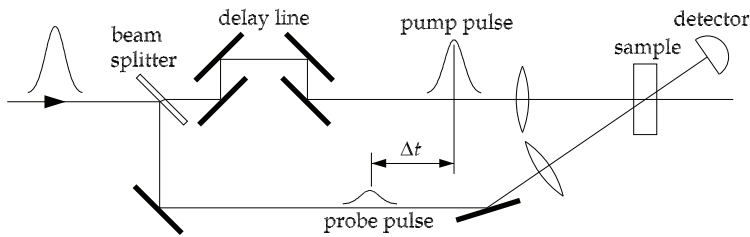


Fig. 1. Scheme of a pump-probe experiment

As an example, consider a two-level sample and the transmitted intensity given by the Beer law (Koechner, 2006; Rullière, 1998):

$$I(\omega, \Delta t) = I_0(\omega) \exp[-\sigma(\omega)n(\Delta t)l] \quad (1)$$

in which  $I_0$  and  $I$  are the incident and transmitted intensities, respectively,  $\sigma(\omega)$  is the absorption cross section at frequency  $\omega$ ,  $l$  is the sample length, and  $n(\Delta t)$  is the ground level population density. An intense resonant pump pulse at  $t = 0$  strongly promotes electrons to the upper level, decreasing the ground level population density  $n(t = 0)$ . If the weaker probe pulse hits the sample at this time  $\Delta t = 0$ , the sample transmittance  $I/I_0$  will be larger than its nominal value, meaning that the absorption coefficient  $\sigma n$  has decreased. As the transmittance measurements are repeated at increasing  $\Delta t$  values, the electrons return to the ground level, and consequently the sample absorption increases. Plotting the transmittance values dependence on  $\Delta t$  leads to the determination of the upper level lifetime. This technique is used when the lifetimes involved are shorter than tens or hundreds of picoseconds, to which solid state detectors do not respond and have low sensitivity. Moreover, the pump or the probe pulse can go through a medium that generates the second or third harmonic, or even a supercontinuum before impinging on the sample, and in this case the effect measured reveals the influence of a frequency on another, providing data to understand its mechanism.

Variations of the technique described can be used to determine the temporal dynamics of many processes in the femtosecond to picosecond time scale. Femtosecond pulses are

known to induce electron heating in metals, and their relaxation, which occurs around 1 ps, can be measured by monitoring the metal reflectivity (Eesley, 1983; Schoenlein et al., 1987) and transmittance temporal evolutions in thin films (Sun et al., 1994). Variations of this technique enable the determination of the characteristic times of the microscopic interactions controlling basic metallic properties like electron transport and superconductivity (Brorson et al., 1990).

Many ultrafast phenomena occur in semiconductors (Rossi & Kuhn, 2002), mainly from the creation of charge carriers in the conduction and valence bands, either by single or multiphotonic absorption of ultrashort pulses (Yariv, 1989). The semiconductor is brought to states far from the thermodynamical equilibrium, and the dynamics of these states relaxations or interaction with electromagnetic radiations can be measured by various pump-probe techniques, with temporal resolutions below 10 fs. Besides transmission and reflection pump-probes measurements, other ones that investigate the carriers evolution bound to the bands, even in coherent states (Leo et al., 1991), can be performed (Axt & Kuhn, 2004).

The dynamics of fast magnetic effects also can be studied using pump-probe techniques. A possible way to do this is to use a polarized pump pulse to optically perturb the magnetization vector of a sample in a magnetic field. After the perturbation, the magnetization precesses in the external field to return to its initial state, modulating the material optical transmission and reflectivity of a polarized probe pulse, allowing the reconstruction of the trajectory of the magnetization vector in 3D space (Andrade et al., 2006; Vomir et al., 2005). Ultrafast spin dynamic can also be determined with similar techniques (Beaurepaire et al., 1996).

In the last decade, the robustness and reliability of ultrashort pulse systems made them valuable tools to pump probe studies of chemical (Zewail, 2000) and biological ultrafast phenomena. Pump-probe techniques have been used to investigate protein dynamics (Pal et al., 2002), intracellular processes such as the dynamics of electrons in photosynthesis (Brixner et al., 2005), and also in photoinitiated DNA degradation mechanisms (Schreier et al., 2007), demonstrating that ultrafast phenomena are essential to life.

Also in the last years, the availability of attosecond ( $10^{-18}$  s) pulses for pump-probe techniques allowed the observation of electronic processes inside atoms (Drescher et al., 2002; Mauritsson et al., 2010; Uiberacker et al., 2007) with temporal resolution in the hundreds of attoseconds. This time-scale is comparable to the revolution time of the electron in the Bohr atom, making possible the experimental investigation of fundamental aspects of quantum mechanics.

Summing it up, pump-probe techniques can use ultrashort pulses, the fastest events ever created by man, to measure and determine the evolution of ultrafast processes in many kinds of materials, molecules or even in internal states of atoms, opening many areas of research and development that a few decades ago were impenetrable to probing. In this context, we expect in the near future to witness the use of attoseconds pulses to probe the dynamics of electrons bound to atoms and investigate fundamental aspects of their wavefunctions.

### 3. Ultrashort pulses nonlinear optics

When light, an electromagnetic oscillating field, impinges on matter its electrons are subjected to the Lorentz force (Jackson, 1999):

$$\mathbf{F} = e(\mathbf{E} + \mathbf{v} \times \mathbf{B}) \quad (2)$$

where  $e$  is the electron charge,  $\mathbf{E}$  and  $\mathbf{B}$  the electric and magnetic fields, and  $\mathbf{v}$  is the electron velocity. Usually the magnetic force is disregarded because the magnetic field amplitude  $B$  is given by the electric field amplitude divided by the speed of light ( $E/c$ ), and the speed  $v$  is small. In this approximation, the electron can be considered to oscillate driven solely by the electric field, and the material response depends on how the electrons trajectories evolve in time. In metals, the electrons are free to follow the excitation field, absorbing its energy in a thin superficial layer and re-emitting it, not allowing electrical fields in its interior. If the electromagnetic field oscillating frequency is above a specific frequency to which the metal free electrons are unable to follow, called the plasma frequency (Jackson, 1999), the metal becomes transparent to the radiation. In a dielectric, the electrons are bound to the atoms by restorative forces, and when an electron is displaced from its equilibrium position by the electric field, an oscillating dipole momentum  $\mathbf{p}(\mathbf{r}, t)$  is induced, therefore emitting electromagnetic radiation at its oscillating frequency (Jackson, 1999). The sum of all dipoles is the material total polarization,  $\mathbf{P}$ , whose spatial components in the frequency domain (Butcher & Cotter, 1990; Shen, 1984) can be written as an expansion in powers of the electric field with coefficients  $\chi^{(n)}$ , called the electrical susceptibilities:

$$P_i = \chi_{ij}^{(1)}(\omega'; \omega_1) E_j(\omega_1) + \chi_{ijk}^{(2)}(\omega''; \omega_1, \omega_2) E_j(\omega_1) E_k(\omega_2) + \chi_{ijkl}^{(3)}(\omega'''; \omega_1, \omega_2, \omega_3) E_j(\omega_1) E_k(\omega_2) E_l(\omega_3) + \dots \quad (3)$$

In expression (3) the right term on the first line represents the linear polarization, given by the product of the instantaneous electrical field component  $E_j(\omega_1)$  oscillating at the frequency  $\omega_1$  and the linear susceptibility tensor  $\chi_{ij}^{(1)}(\omega'; \omega_1)$ , in which the indices indicate the spatial coordinates ( $i, j = x, y, z$ ). In this susceptibility notation, the polarization oscillation frequency is the one at the left of the semicolon, and it is the sum of the electrical fields frequencies at the right. These frequencies can be either positive or negative, and in this case ( $\omega_i < 0$ ), the corresponding electrical field complex conjugate is used (Butcher & Cotter, 1990). When the exciting field amplitude is small and the electron oscillation is harmonic, the polarization is proportional to the field and oscillates at its frequency ( $\omega' = \omega_1$ ). The arising effects, such as the refractive index and the absorption, are linear on the field and do not depend on its intensity (Shen, 1984).

As the light intensity and the electrical field amplitude increase, the electrons oscillations became anharmonic and higher orders of the expansion have to be added to the total polarization (Butcher & Cotter, 1990; Shen, 1984), as exemplified by the second and third orders nonlinear polarizations on the bottom line of expression (3). Each nonlinear polarization is the product of a nonlinear susceptibility tensor and various exciting fields. As long as the electrons stay bound to the atoms, this description can be used, and many nonlinear phenomena arise from the different nonlinear susceptibilities spatial components and frequencies combinations. As the nonlinear polarizations grow, the total polarization oscillates in various frequencies simultaneously, and consequently the emission will be the superposition of monochromatic fields.

Regarding the mechanisms that generate nonlinear optics phenomena, the main ones are electronic redistribution and molecular reorientation (electronic configurations changes),

and thermal and electrostriction processes that modify the material density (Boyd, 2008). All these processes can be described by the formalism presented, but only the phenomena resulting from electronic redistribution will be discussed here once the other processes are slow and do not respond to ultrashort pulses.

Ultrashort pulses are prone to generate nonlinear effects due to the high powers and intensities readily attainable. Even a few nanojoules of energy in a 50 femtoseconds pulse, straightforwardly generated by ultrafast lasers, produce tens of kilowatts of peak power, and can be easily focused in a spot with a radius of 20  $\mu\text{m}$ , resulting in intensities on the order of  $\text{GW}/\text{cm}^2$ . From the Poynting vector it can be shown (Born & Wolf, 1999) that the electric field amplitude,  $E$ , is related to the intensity,  $I$ , by:

$$E(\text{V}/\text{cm}) = 27.45 \sqrt{\frac{I(\text{W}/\text{cm}^2)}{n_0}} \quad (4)$$

where  $n_0$  is the refractive index of the medium. Intensities of  $\text{GW}/\text{cm}^2$  correspond to electric fields of about  $10^6$   $\text{V}/\text{cm}$ , which is on the order of 1% of the electric field binding electrons to molecules, meaning that at these intensities the contribution of the higher order electric field terms of expression (3) start to be relevant to the polarization. Higher intensities that increase the nonlinear effects can be obtained from CPA systems, extended cavity lasers and cavity dump techniques.

The first susceptibility to generate nonlinear effects is the second order one,  $\chi^{(2)}$ . If one of the fields is zero, the susceptibility  $\chi^{(2)}(\omega; 0, \omega)$  describes the Pockels effect (Fowles, 1989), in which a static (DC) electric field applied to a medium rotates the polarization of a field at the optical frequency  $\omega$  going through this medium. Choosing adequately the medium length and the DC field amplitude, it is possible to rotate by 90 degrees the polarization of an ultrashort pulse at  $\omega$ , and a polarizer placed after the medium will transmit or block the pulse depending if the DC field is on or off. This arrangement, called a Pockels Cell, is commonly used as a fast optical switch in ultrashort laser pulse systems.

The second order susceptibility component  $\chi^{(2)}(\omega; \pm\omega_1, \pm\omega_2)$  also describes a class of phenomena, called sum-frequency generation, in which two photons from different laser beams are mixed to create a third one (Boyd, 2008). As already explained, when a negative frequency is used, the complex conjugate corresponding field is used in the polarization expression, so in the particular case that  $\omega_1$  is positive and  $\omega_2$  is negative, the second order polarization is:

$$\begin{aligned} P_i^{(2)} &= \chi_{ijk}^{(2)}(\omega_1 - \omega_2; \omega_1, -\omega_2) E_{0j}(\mathbf{r}, t) e^{-i\omega_1 t} \left[ E_{0k}(\mathbf{r}, t) e^{-i\omega_2 t} \right]^* \\ &= \chi_{ijk}^{(2)}(\omega_1 - \omega_2; \omega_1, -\omega_2) E_{0j}(\mathbf{r}, t) E_{0k}^*(\mathbf{r}, t) e^{-i(\omega_1 - \omega_2)t} \end{aligned} \quad (5)$$

where  $E_0(\mathbf{r}, t)$  is the electric field temporal and spatial envelope (Jackson, 1999). The negative frequency  $-\omega_2$  means that this field carries a phase of  $\pi$  relative to  $\omega_1$ , so the polarization oscillates at the difference frequency, as evidenced in equation (5), and a photon is emitted with energy  $\hbar(\omega_1 - \omega_2)$ . The exciting photons are not resonant with the transparent medium, and virtual electronic levels are involved (Yariv, 1989). Note that when two photons, at frequencies  $\omega_1$  and  $\omega_2$  interact with a nonzero second order susceptibility, both the frequencies  $\omega_1 - \omega_2$  and  $\omega_2 - \omega_1$  are simultaneously generated, and a negative values means  $\pi$  added to the phase. An immediate application of sum-frequency generation for ultrashort

pulses is the Optical Parametric Amplification (OPA) (Yariv & Yeh, 2007), in which a powerful pump beam at  $\omega_p$  transfers energy to two beams at lower frequencies  $\omega_i$  and  $\omega_s$ , with  $\omega_p = \omega_i + \omega_s$ , where i and s stand for idler and signal, respectively. The susceptibility describing it is  $\chi^{(2)}(\omega_s; \omega_i, \omega_p)$ , determining how the energy moves back and forth between the three beams, and fine-tuning the phase matching conditions in the gain medium makes it possible to control the resulting amount of energy at each frequency, so a weak idler beam can be used to induce energy transfer from the pump to generate a powerful signal beam. This effect is used to extend ultrashort pulses to wavelengths differing from the main oscillator one, increasing the wavelength range of ultrashort pulses available for spectroscopic and other applications (Laenen et al., 1993). The OPA also can be used to amplify ultrashort pulses into the petawatt regime by the Optical Parametric CPA (OPCPA) technique (Dubietis et al., 2006). The pump beam and the ultrashort pulse to be amplified interact in a gain medium, generating an idler one; adjusting the phase matching conditions the idler beam intensity can be minimized and the signal beam amplified (Cerullo & De Silvestri, 2003). The gain medium is not resonant with either  $\omega_p$ ,  $\omega_i$  or  $\omega_s$ , so no energy is absorbed by it, and thermal problems that are known to limit the amplification performance (Koechner, 2006) are minimized; moreover, with the appropriate parameters selection, the gain medium bandwidth can be extended to all the gain medium transparency window.

A special case of the sum-frequency generation, called Second Harmonic Generation (SHG) or frequency doubling, occurs in the degenerate case when  $\omega_1 = \omega_2 = \omega$  (Boyd, 2008). In this situation, governed by  $\chi^{(2)}(2\omega; \omega, \omega)$ , in which only one laser beam is used, the polarization oscillates at twice the exciting field frequency, emitting light with half of the excitation wavelength. The SHG can also be understood as the absorption of two photons with energy  $\hbar\omega$  followed by the emission of a photon with energy  $2\hbar\omega$ , since energy has to be conserved. Homogeneous media, like gases or glasses, are not capable of SHG. This happens because the second order nonlinear polarization is  $P^{(2)} = \chi^{(2)}E \cdot E$ , and in a homogeneous surrounding, an inversion of the electrical field must result in an inversion of the polarization:  $-P^{(2)} = \chi^{(2)}(-E) \cdot (-E) = \chi^{(2)}E \cdot E$ . This is only possible if  $\chi^{(2)} = 0$ . The SHG signal increases quadratically with the excitation intensity, and generates ultrashort pulses at new wavelengths that can be used in experiments such as the pump-probe ones already mentioned in the previous section.

The SHG can be used to measure the temporal duration of ultrashort pulses by a technique known as second harmonic autocorrelation. As mentioned in the beginning of section 2, to measure an event, a faster phenomenon is needed, and since that there are no man-made events shorter than an ultrashort pulse to measure it, the pulse itself has to be used to determine its own duration. This is made possible by the use of a Michelson interferometer (Träger, 2007) and a SHG crystal placed at the crossing of the two beams, as shown in Fig. 2. The pulse to be measured is split in two copies by the beam splitter (BS), and each copy travels along a different arm. The mirror M in the delay line can be moved back and forth in order to control the temporal superposition of the pulse replicas at the SHG crystal, and when these arrive at the crystal delayed by more than the pulse duration there is no SHG; as the mirror is moved and the pulses overlap in time, a SHG beam proportional to the time superposition (autocorrelation signal) is produced at the bisecting angle defined by the two beams oscillating at  $\omega$ , once momentum has to be conserved. The autocorrelation signal reaches its maximum when the delay between the pulses is zero. This technique generates a symmetrical autocorrelation trace that is proportional to the delay line position, and a slow

detector can be used to measure the second harmonic signal generated by a train of pulses. The deconvolution of the autocorrelation signal, taking into account that a displacement of 1  $\mu\text{m}$  corresponds to 3.3 fs, determines the pulse duration (Trebino, 2000).

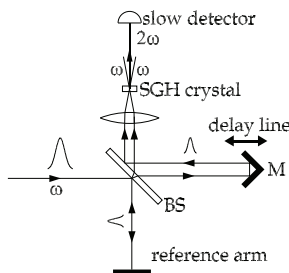


Fig. 2. Second order autocorrelation scheme. The thicker beam originating at the SHG crystal is the autocorrelation signal.

The second order autocorrelation is the most used technique to determine the duration of ultrashort pulses, although it presents some problems, such as ambiguities in the pulse temporal shape and no retrieval of any information about the pulse phase (Trebino, 2000). In order to overcome these problems and to completely determine the pulse temporal and spectral shapes and phases without ambiguities, other techniques, such as Frequency Resolved Optical Gating (FROG) (Kane & Trebino, 1993; Trebino, 2000) and Spectral Phase Interferometry for Direct Electric-field Reconstruction (SPIDER) (Gallmann et al., 1999) were created, allowing the measurement of pulses below 5 femtoseconds. Variations of these methods, in which an ultrashort pulse whose duration is known is used to generate the second harmonic interacting with a unknown pulse can be easily implemented, and are called cross-correlation techniques (Trebino, 2000). The deconvolution of the SHG signal with knowledge of one of the pulses determines the unknown one.

The two photon induced fluorescence is a variation of the SHG technique in which two photons are absorbed to a resonant level of a molecule, called fluorophore, and a more energetic photon is emitted (Peticolas et al., 1963). Although the process, differently from the previous one described, is resonant with two photons, it can also be described by the  $\chi^{(2)}(2\omega; \omega, \omega)$  of the fluorophore. This effect can be used in a technique called two-photon scanning microscopy (Denk et al., 1990; Zipfel et al., 2003), in which a fluorophore that is transparent to the ultrashort pulses, but exhibits two-photon absorption for the same pulses, is used to image microscopic samples, mainly biological ones (Konig, 2000; Xu et al., 1996). A strongly focused infrared laser beam is used, and the two-photon fluorescence is confined to the focus region due to the nonlinear dependence of the emission on the excitation intensity. The laser beam is scanned across the sample, mapping the fluorophores with 3D high-resolution (Muller et al., 1998). Besides using naturally occurring fluorophores, molecules can be engineered to have high values of  $\chi^{(2)}(2\omega; \omega, \omega)$  (Albota et al., 1998) and to bind to specific proteins so structures can be imaged unambiguously. The high precision obtained by the nonlinear effect at the vicinity of the focus spot can also be used to manufacture microstructures using two-photon polymerization. In this process, instead of using a fluorophore, a monomer that photopolymerizes under ultraviolet light and is transparent to the infrared is illuminated by an infrared laser. In this material the laser beam can be focused in deep regions, and the polymerization is initiated by two-photon absorption in a small region surrounding the beam focus, enabling the fabrication of three-dimensional structures



(Cumpston et al., 1999; Maruo et al., 1997). Both the two-photon microscopy and two-photon polymerization take advantage of the transparency of the medium for the fundamental wavelength, which allows the laser propagation inside the medium with minimal losses, and the nonthermal character of the ultrashort pulses interaction, that does not heat the samples, avoiding thermal effects that can modify the samples properties.

Moving to the third order susceptibility, some phenomena are extensions of the second order ones, such as the four wave mixing governed by  $\chi^{(3)}(\omega; \pm\omega_1, \pm\omega_2, \pm\omega_3)$  (Träger, 2007), in which three photons at frequencies  $\omega_1, \omega_2$  and  $\omega_3$  interact to generate a fourth one. Depending on the particular frequencies involved several phenomena as Coherent Anti-Stokes Raman Scattering (CARS) (Zumbusch et al., 1999), Degenerate Four Wave Mixing (DFWM) (Joo & Albrecht, 1993), Optical Phase Conjugation (Yau et al., 1996), Stimulated Brillouin Scattering, among others (Yariv & Yeh, 2007), can be generated. In the degenerate case for a single beam the third harmonic, described by  $\chi^{(3)}(3\omega; \omega, \omega, \omega)$ , is generated, and some applications are extensions of the second order ones (Träger, 2007).

Another class of phenomena emerge when the effects arising from the third order susceptibility  $\chi^{(3)}(\omega; \omega, \omega, -\omega)$  come into play for a single beam. Under these conditions the third-order polarization is

$$P_i^{(3)} = \chi_{ijj}^{(3)}(\omega; \omega, \omega, -\omega) E_{0j}(t) e^{-i\omega t} E_{0j}(t) e^{-i\omega t} [E_{0j}(t) e^{-i\omega t}]^* = \chi_{ijj}^{(3)}(\omega; \omega, \omega, -\omega) e^{-i\omega t} E_{0j}(t) I_0(t) \quad (6)$$

where  $I_0(t) = E_{0j}(t) \cdot [E_{0j}(t)]^*$  is the beam intensity. Expression (6) shows that the polarization oscillates at the same frequency of the exciting beam and it is proportional to its intensity, and this implies (Butcher & Cotter, 1990; Shen, 1984) that the total refractive index of the material,  $n$ , must have a dependence on the pulse intensity according to:

$$n(\mathbf{r}, t) = n_0 + n_2 I(\mathbf{r}, t) \quad (7)$$

where  $n_0$  is the linear (usual) refractive index of the material,  $I$  is the pulse intensity and  $n_2$  is the nonlinear refractive index given by  $n_2 = \alpha \text{Re}[\chi^{(3)}(\omega; \omega, \omega, -\omega)]$ , where the constant  $\alpha$  depends on the unit systems considered (Butcher & Cotter, 1990). Inserting equation (7) in the expression of a plane wave  $E_0 \exp[i(knx - \omega t)]$  propagating in the  $x$  direction results in:

$$E(\mathbf{r}, t) = E_0(t) \exp[i(kn_0 x - \omega t)] \exp[ikn_2 I(\mathbf{r}, t)x] \quad (8)$$

where  $k = 2\pi/\lambda$  is the plane wave wavenumber. The second exponential represents the nonlinear effects that can result from the spatial and temporal distributions of the intensity. Considering a laser beam with a spatial Gaussian intensity distribution (Yariv, 1989) propagating in the  $x$  direction, the intensity reaches its maximum in the optical axis, and decays exponentially in the plane perpendicular to the propagation, and the spatial profile of the total refractive index follow this distribution. If  $n_2$  is positive, the beam center goes through a medium with a longer optical path than its borders, similarly to a convergent lens, and the beam is focused. This effect is known either as Kerr effect or self-focusing (or self-defocusing if  $n_2$  is negative) (Shen, 1984), and is a self-effect in which the beam characteristics determine its propagation through a nonlinear medium. The self-focusing effect is used in many ultrashort lasers as the Kerr-Lens Mode-Locking mechanism (Haus et al., 1992) responsible to decrease the resonator losses for shorter pulses (Koechner, 2006), providing a robust and stable way to passively generate the shortest pulses possible directly from laser oscillators.

Based on the self-focusing effect, a simple technique was devised to measure the value of  $n_2$ , and consequently of  $\chi^{(3)}(\omega; \omega, \omega, -\omega)$ . This technique, called Z-Scan (Sheik-Bahae et al., 1990), consists in scanning a nonlinear sample across the waist of a focused beam, measuring the beam transmittance through an iris in the far-field as a function of its position. For a sample with positive  $n_2$  self-focusing occurs, and when the sample is before the beam waist the tighter focusing results in a higher divergence, decreasing the transmittance through the iris; conversely, when the sample is after the waist, the self focusing reduces the beam divergence and the transmittance through the iris increases. These effects produce a peak-valley curve of the transmittance dependence on the sample position, and measuring the peak-valley signal variation immediately determines the third order nonlinear susceptibility. The Z-Scan technique quickly became widely used due to its simplicity, good sensitivity and the capability of measuring the electronic nonlinearities of solid and liquid materials. Many variations were introduced in the Z-Scan technique in order to increase its sensitivity (Kershaw, 1995), to resolve the time-scales of the self-focusing process with a pump-probe measurement (Ma et al., 1991), to measure dispersion of the nonlinearity (Balu et al., 2004) and to consolidate it as the most used technique to measure odd-orders nonlinearities (Zhan et al., 2002) in crystals, glasses, polymers and solutions among other kinds of samples. Alternatively, performing a Z-Scan measurement without the iris with all the beam energy impinging on a detector, the nonlinear absorption, corresponding to the imaginary part of  $\chi^{(3)}(\omega; \omega, \omega, -\omega)$  is measured (Sheik-Bahae et al., 1990), providing more information about the electronic processes occurring inside the sample under study.

Taking into account now the temporal aspects of expression (8), consider a temporally symmetric pulse centered at the temporal origin with a temporal distribution  $I(t)$  that can be expanded around  $t = 0$ . The expansion can be written as  $I(t) = I_0 + \beta t$ , with  $\beta = \partial I(t)/\partial t$ , and its substitution in expression (8) results in:

$$E(\mathbf{r}, t) = E_0(t) \exp[ik(n_0 + n_2 I_0)x] \exp[-i(\omega - kn_2 \beta x)t] \quad (9)$$

As can be seen in the second exponential of expression (9), the pulse carrier frequency is shifted by  $kn_2 \beta x$ , meaning that before the pulse peak, lower frequencies are generated, and after it higher frequencies are created. This Phenomena is called Self-Phase Modulation (SPM) (Yariv, 1989) and is responsible for broadening the spectrum inside laser resonators for the generation of ultrashort pulses (Koechner, 2006). Without SPM the gain media would not generate enough gain to maintain the bandwidth needed, by Fourier Transform, to generate ultrashort pulses. In some specially designed resonators it is nowadays possible to generate octave-spanning spectra that support pulses shorter than 5 fs (Ell et al., 2001), and these ultra-broadband pulses are used to synthesize optical combs (Holzwarth et al., 2000; Ye & Cundiff, 2005). These optical combs are frequency synthesizers with uncertainties below  $10^{-15}$  that are being used as time standards replacing atomic clocks (Takamoto et al., 2005), and to absolutely measure optical frequencies (Reichert et al., 1999; Udem et al., 2001). These combs are also used to stabilize the phase between the pulse carrier frequency and its envelope to a few milirads, corresponding to tens of attoseconds (Telle et al., 1999).

The SPM is also one of the main mechanism, along with wave mixing and harmonics generation, responsible by the generation of white light supercontinuum (Alfano, 2006) that can support shorter than 5 fs pulses with energies at the few milijoule level (Bohman et al., 2010). These supercontinuum extend the ultrashort pulses to new wavelengths, expanding the range of applications such as pump-probe ones, keeping the ultrafast aspect of the interaction with matter.

There are many other nonlinear phenomena acting in the time interval ranging from a few picoseconds down to the attosecond region, and only a few important ones were described here. Whole books were written on this subject (Diels & Rudolph, 2006; Hannaford, 2005; Rullière, 1998; Träger, 2007) and the reader interested in a deeper understanding is encouraged to resort to these references.

#### 4. Ionization by ultrashort pulses

When the intensity of ultrashort pulses reaching the electrons in a material is high enough to exceed the electric field binding the electrons, ionization takes place. Depending on the density of free electrons generated in solid samples, ablation can occur on the material surface, or other phenomena can happen inside the sample.

Ultrashort pulse laser ablation of solids is due to an electron avalanche induced breakdown process (Bloembergen, 1974; Du et al., 1994) that occurs when seed electrons are accelerated in the laser field, exponentially generating free electrons by collisions. The breakdown takes place when the plasma originated by the avalanche electrons reaches a critical density and transfers energy to lattice ions, which expand away from the surface after the pulse has finished. In metals, the seed electrons are always present (conduction band free electrons), and in dielectrics and semiconductors they are excited from the valence to the conduction band by the pulse leading edge, either by multiphotonic ionization (Kautek et al., 1996; Perry et al., 1999) or by tunneling induced by the laser field (Keldysh, 1965; Lenzner et al., 1998). Although the seed electrons have dissimilar origins in different classes of materials, a metallization occurs in dielectrics and semiconductors after they are produced, and the avalanche evolves deterministically in time (Bass & Fradin, 1973; Du et al., 1994; Joglekar et al., 2003) in the same way in all solids, that behave like metals (Gamaly et al., 2002; Nolte et al., 1997). These mechanisms confer a nonselective characteristic to the ultrashort pulse ablation, and the intensity ablation threshold of a material,  $I_{thr}$ , is the only parameter relevant to the etching process. The established method (Liu, 1982) to determine the ultrashort pulses ablation threshold of a given material consists in ablating this material with a TEM<sub>00</sub> Gaussian beam at various intensities, and then measuring each ablation area diameter. From these data the ablation threshold is determined. A few years ago members of our group analytically described (Samad & Vieira, 2006) and introduced an alternative simpler method to measure the ultrashort pulses ablation threshold of solid samples, based in the very precise definition of the etching region resulting from the nonlinear character of the ultrashort pulse ablation together with the almost inexistent lateral heat diffusion. The method consists in moving the sample diagonally across the waist of a focused ultrashort pulses laser beam, etching the profile shown in Fig. 3 in its surface.

It can be easily shown that the maximum transversal dimension of the etched profile,  $\rho_{max}$ , is related to the ablation threshold,  $I_{thr}$ , by:

$$\rho_{max} = 0.342 \sqrt{\frac{P_0}{I_{thr}}} \quad (10)$$

where  $P_0$  is the ultrashort pulse power, readily measurable in a laser laboratory. The values of ablation threshold measured with this technique agree with the ones given by the traditional method (Freitas et al., 2010; Samad et al., 2008).

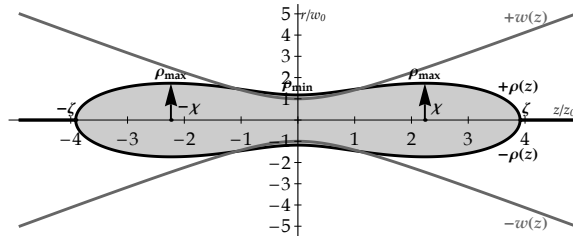


Fig. 3. Ablation profile etched in the surface of a sample by the diagonal scan method. The laser beam size is indicated by the curves labeled  $\pm w(z)$ , and the horizontal and vertical axes are normalized by the beam confocal parameter  $z_0$  and beamwaist  $w_0$ , respectively.

Once the ablation threshold is known, ultrashort pulses can be used in a variety of materials ranging from transparent dielectrics (Gattass & Mazur, 2008) to metals (Dumitru et al., 2002; Itoh et al., 2006; Ke et al., 2005; Shirk & Molian, 1998), semiconductors (Nayak et al., 2007) and polymers (Baudach et al., 2000), to drill holes and to machine the material in a controlled way to modify its surface (Zoubir et al., 2003), create microstructures (Kruger & Kautek, 1999; Sugioka et al., 2005) and microchannels (Gomez et al., 2005). Due to the nonthermal character of the ablation, the heat affected zone is minimized, and can be brought to be almost nonexistent, allowing the use of ultrafast pulses to perform surgeries (Vogel et al., 2005; Vogel & Venugopalan, 2003) like neuron severing without damaging neighboring living tissues (Chung et al., 2006) and corneal cutting as high precision scalpels (Juhasz et al., 1999), among other application in biological samples and tissues (Braun et al., 2008).

At lower intensities than those needed for ablation, the free electrons density does not reach the value needed to produce the Coulombic explosion, but the localized excess of free electrons can be enough to promote modifications in the material bulk such as permanent changes in its refractive index (Davis et al., 1996) and the creation of defects like color centers (Courrol et al., 2004). We suggest that the defects creation mechanism begins with electrons being teared out from anions off the structure by multiphotonic or impact ionization, leaving a neutral atom behind. This neutral atom is no longer held in its position by the surrounding electric field, so it can be moved from its site by a collision with an electron oscillating in the laser field, and when this occurs, an electron can be trapped in the potential of the now empty position, creating a color center (Courrol et al., 2004; Gellermann, 1991). Alternatively, electrons can be trapped in the UV or valence bands, modifying electronic properties of the material, and consequently changing its refractive index. These processes and similar ones happen in glasses (Courrol et al., 2008), crystals (Martynovich et al., 2008; Orlando et al., 2010), and polymers (Samad et al., 2010), and when these defects are created with spatial control inside the material, structures such as waveguides (Nolte et al., 2003), diffraction gratings (Hirao & Miura, 1998) and photonic devices (Florea & Winick, 2003; Minoshima et al., 2001) can be manufactured by the ultrashort pulses. Once again, all these processes benefit from the minimized heat generation that preserves the material properties in the surroundings of the created structures.

## 5. Plasma, high harmonic generation and attosecond pulses

When ultrashort pulses interact with a material at intensities higher than those needed for ablation, the excess energy delivered by the pulses accelerates electrons, produce higher

ionization states and excites the ions to upper energy levels, forming a light emitting plasma (Gibbon & Forster, 1996) whose properties are essentially controlled by the laser pulse characteristics.

Ultrashort pulses lasting 30 fs with modest energies under 1 mJ can be easily focused on sample surfaces to intensities above  $10^{15}$  W/cm<sup>2</sup>, generating plasmas whose emission is used to determine the elements of the sample in a technique called femtosecond Laser Induced Breakdown Spectroscopy (fs-LIBS) (Le Drogoff et al., 2001). The traditional LIBS technique (Cremers & Radziemski, 2006) relies on the characteristic light emission of each element, being used as an analytical tool for qualitative and quantitative analysis of the sample composition, and is performed with nanosecond pulses that promote ablation through thermal processes and then excite the ejected atoms by multiphotonic absorption. The thermal character of the ablation creates a dependence of the LIBS signal on the type of material under study, demanding the determination of many calibration curves. In fs-LIBS the nonselective character of the ultrashort pulses ablation makes the technique almost insensitive to the material under study, so less calibration curves must be created, and almost any solid material can be studied, including biological (Baudalet et al., 2006; Samek et al., 2006; Santos et al., 2008) and archeological samples, and works of art in order to determine the materials used by their artists (Svanberg, 2008). Because the plasma is formed by focused optical radiation, LIBS can also be used to interrogate samples remotely by stand-off analysis.

The plasma formation capability of ultrashort pulses can also be used in conjunction with LIDAR systems (Weitkamp, 2005) to investigate the atmosphere composition. For this, terawatt pulses are sent into the atmosphere with negative dispersion, and are compressed by the atmosphere positive dispersion until self-focusing occurs, leading to an abrupt intensity increase that leads to breakdown and plasma formation. The plasma introduces a defocusing effect that, under the right conditions, balance the self-focusing, channeling the plasma into filaments that can propagate through hundreds of meters (Kasparian et al., 2003), placing an intense light source in the sky. The plasma emission is collected by a ground based telescope, providing information about the atmospheric composition and transmittance. More recently, this plasma formation processes have been under study to induce water condensation in the atmosphere as a rain starting mechanism (Rohwetter et al., 2010), and to control lightning (Kasparian et al., 2010).

When plasmas are generated in gases at low pressures, the ionized electrons can be accelerated to high energies before colliding with ions. This mechanism is known as three-step model, in which the ultrashort pulse initially ionizes an atom by either tunneling or multiphotonic absorption (Kautek et al., 1996; Keldysh, 1965; Miyazaki & Takada, 1995), then the electron is accelerated away by the oscillating electric field that brings it back to collide with the ion, emitting x-rays as its kinetic energy is converted to electromagnetic radiation (Daido, 2002). Depending on the experimental details the x-rays generated can be generated in a coherent way, originating a beam with laser characteristics. These x-rays obey selection rules that determine the wavelengths generated, that can be understood as high harmonics of the fundamental ultrashort pulse (L'Huillier & Balcou, 1993; Lewenstein et al., 1994). Among other applications, these x-ray pulses can be used for high resolution imaging (Chao et al., 2005) in the water window (Chang et al., 1997; Gibson et al., 2003; Spielmann et al., 1997) of biological systems, and also of biomolecules and proteins (Neutze et al., 2000).

Assuming a pulse with a Gaussian temporal profile without generality loss, and referring to equation (3), it can be seen that when an harmonic is generated by the  $n$ -order polarization,

the harmonic pulse is shortened by a factor  $n^{1/2}$  (Shen, 1984). If the harmonic order is sufficiently high, the harmonic pulse can be generated with durations below a hundred attoseconds (Krausz & Ivanov, 2009; Tang & Chen, 2010) in the x-ray spectral region. This is the timescale of the electrons movements in atomic and molecular orbitals (Corkum & Krausz, 2007), and these pulses can be used in pump-probe experiments to directly observe electron tunneling in the dynamics of ionization (Kling & Vrakking, 2008; Uiberacker et al., 2007) and to control ionization processes (Johnsson et al., 2007). To deal with pulses in the attosecond time scale, new experimental tools and techniques are being developed to control and measure these pulses, like x-ray photoemission, cross correlation of light and x-rays (Hentschel et al., 2001). Today is possible to obtain information on the amplitude and phase of electronic wavefunctions using attosecond pulses.

## 6. Relativistic optics and high field science

When intensities on the order of  $10^{18}$  W/cm<sup>2</sup> are reached and the corresponding electric field, given by expression (4), is an order of magnitude greater than the electric field in the Bohr atom, relativistic nonlinear effects start to come into play. At these intensities, the electron quivering motion on an ultrashort pulse reaches relativistic velocities, and the magnetic term on the Lorentz force, equation (2), has to be taken into account since  $v/c$  approaches the unity and the magnetic force is comparable to the electric one. In this situation the magnetic force pushes the electron in the forward direction, along with the Poynting vector. A direct consequence of this is wakefield generation, a relativistic optical rectification, in which the longitudinal field effects could be as large as the transverse ones (Umstadter et al., 1996). The electrons accelerated in this configuration are originated from a plasma, and create a strong static field that pull the ions left behind, accelerating them. Protons can be accelerated to energies over a GeV, in what are being called laser driven particle accelerators (Bulanov et al., 2010; Maksimchuk et al., 2000).

At these high intensities, relativistic phenomena analogous to nonlinear ones appear, as relativistic focusing in which the focusing is due to the relativistic electron mass increase (Monot et al., 1995), relativistic transparency, nonlinear modulation and multiple harmonic generation, and strong coupling to matter and other fields also take place (Mourou et al., 2006).

At very high intensities the light pressure can be used to inertially confine targets that will reach densities and temperatures high enough to obtain fast ignition in a laser driven atomic fusion process (Moses et al., 2009). The densities and temperatures expected are almost an order of magnitude higher than those at the center of the sun, allowing the accomplishment of experimental astrophysics.

Nowadays, many laboratories are working to increase the power generated by ultrashort lasers, and ways to focus it to ultimately reach intensities close to  $10^{29}$  W/cm<sup>2</sup>, the Schwinger limit (Bulanov et al., 2003; Schwinger, 1951), at which electron positron pairs can be created directly from the quantum vacuum as a consequence of its polarization by the electromagnetic field.

## 7. Conclusions

Applications of ultrashort pulses were presented and discussed here. Some basic optical mechanisms that permeate many applications were outlined to create a basis for the reader

to expand his knowledge on the field of ultrashort pulses. Although the discussions of the many applications are far from complete once almost each topic presented here can be expanded to fill a complete book, updated references in the form of books, reviews and cornerstone papers were given, and the reader is encouraged to read them to obtain a deeper comprehension on the subjacent mechanisms and resulting phenomena.

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