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Generation and diagnostics of femtosecond light pulses

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We review basic techniques for generation of ultra-short light pulses. In particular, we discuss different methods of mode-locking, pulse amplification and frequency conversion methods based on the 2nd and 3rd order nonlinear processes. Basic methods for pulse characterisation are also discussed.

Keywords: femtosecond laser pulses, ultra-short pulse characterisation.

1. Introduction

During the last decade, developments in laser mode-locking techniques led to generation of pulses shorter than 6 fs (1 fs = 10^{-15} s) directly from Ti:Sapphire laser [1]. By employing pulse compression techniques one can achieve even shorter pulses with the current record at less than 5 fs level [2,3]. Impressive progress in this field during the 70s and 80s is now followed by maturing of the technology. Thus, while the early femtosecond systems were based on organic dyes as laser materials, with an advent of femtosecond Ti:Sapphire [4] and other vibronic lasers in the early 90s a solid state technology is now available. Currently, low power systems operating in 20–30 fs range are routine and commercial systems with sub-10 fs capabilities are on the market. Even more impressive is the progress in ultra-short pulse amplification and frequency conversion. Development of the chirped pulse amplification technique coupled with harmonic generation and parametric devices enables generation of sub-100 fs pulses anywhere from UV to mid-infrared. At the other end of the spectrum THz pulses span the frequency range from radio frequencies up to tens of THz. It is only natural that the ever improving generation techniques lead to numerous applications in both basic research and technology, e.g., time resolved spectroscopy, femtochemistry, imaging, telecommunications, etc. The aim of this contribution is to review and summarise the current state of the field. Although the most rapid growth in the femtosecond technology is probably over there are still important problems to be solved and developments to be accomplished.

This contribution is organised as follows. In Section 2 we present basic techniques for generation of femtosecond light pulses. Section 3 is devoted to the methods used to characterise those pulses. Section 4 contains some remarks on the prospects of the field.

2. Generation of femtosecond light pulses

The history of ultra-short laser pulse generation (see Fig. 1) dates back to the 1960-ties when mode-locking in He-Ne laser with sub-ns pulses was reported for the first time [5]. Since then a steady progress in the field resulted in continuous redefining of the notion "ultra-short laser pulse". While in the early 1970-ties it meant pulses with duration of a few ps, by mid-1980-ties ultra-short meant sub-100 fs and today the same term applies to the pulses with duration of just a few fs. Recently sub-femtosecond transients in high harmonic generation in argon were observed and interpreted as attosecond (1 attosecond = 10^{-18} s) pulses of radiation [6].

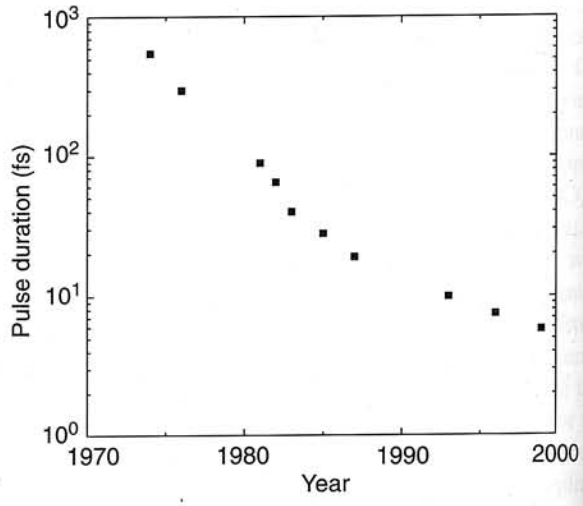

Fig. 1. History of the ultra-short light pulses obtained directly from lasers.

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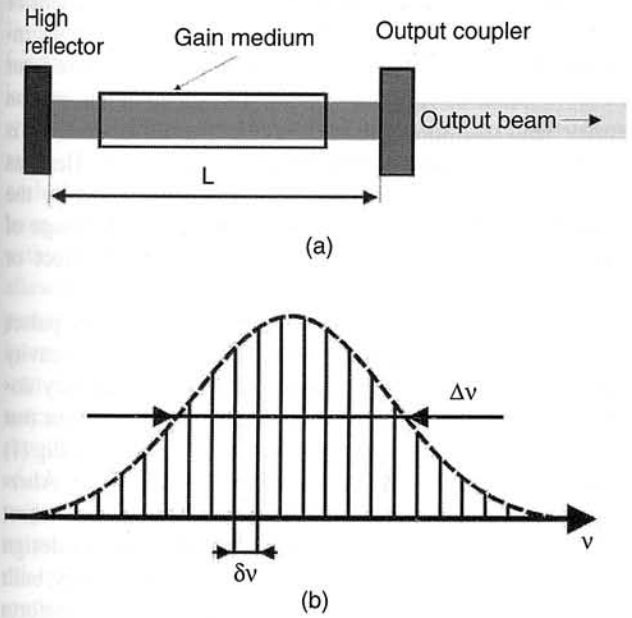

Fig. 2. A laser (a) and its spectrum (b).

Ultra-short pulse generation in all lasers is based on the phenomenon called mode-locking. The underlying idea is very simple. Given a gain medium with sufficiently broad gain spectrum one can, in principle, make the laser oscillate at many longitudinal modes simultaneously. If we ignore the dispersion of the laser cavity then the modes are evenly spaced in frequency as illustrated in Fig. 2. The laser output beam is, in this case, a coherent superposition of many basically monochromatic waves corresponding to individual cavity modes and its intensity is given by

$$I(t) = \left| \sum_k E_k \cos(2\pi \nu_k + \varphi_k) \right|^2 \qquad (1)$$

where E_k is the amplitude of the k-th mode and ν_k and φ_k are its frequency and phase, respectively. The result of such a superposition dramatically depends on relative phases of the modes (see Fig. 3). One can consider two limiting cases. When the modes are not coupled at all, their phases are independent and totally random. In this case, the laser intensity fluctuates in time with peak intensity scaling like the number of oscillating modes. On the other extreme one can consider a case when the modes are coupled by a certain mechanism. The coupling leads to correlations between the phases of individual modes. Let's assume, for simplicity, that the coupling between modes is so strong that all the phases are identical. Then the result of the superposition is a regular train of pulses with the repetition rate equal to the inverse of the cavity round-trip time and pulse duration inversely proportional to the bandwidth of the gain medium. In this case the peak intensity scales like the square of the number of modes. Our model of mode-locking is oversimplified in two respects. First, we

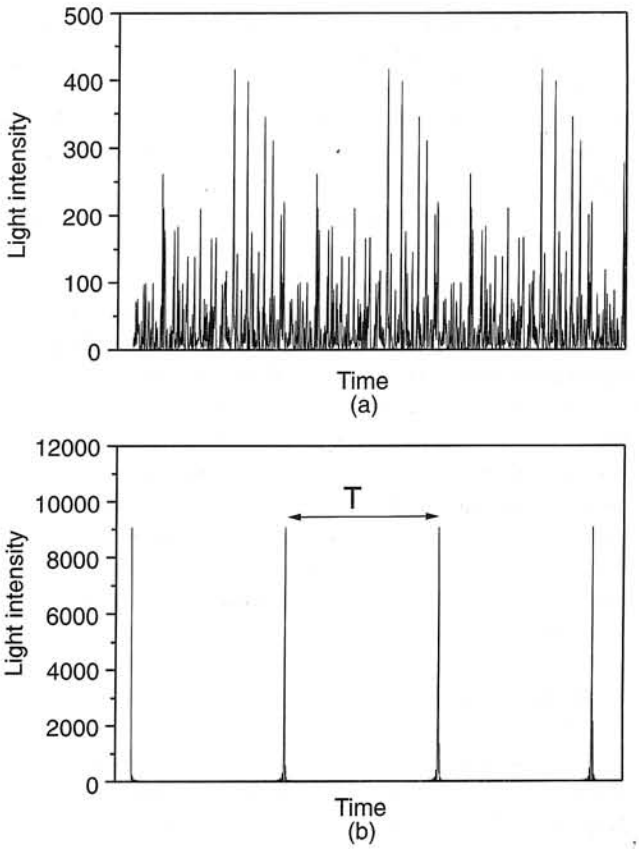

Fig. 3. Laser intensity versus time. The plots have been generated with Eq. (1). In both cases 100 modes of equal amplitudes have been assumed. In (a) the phases of the modes are random and in (b) the phases are all equal zero.

have assumed that the phases of all the modes are perfectly correlated (in fact we have assumed that they are identical). We have also ignored the dispersion of the gain medium and other components of the laser cavity. We will briefly discuss both effects.

Imperfect mode-locking implies that coupling between modes is not strong enough to assure constant phase relations. One can think of it as a jitter imposed on the phases of individual modes which results in temporal broadening of the output pulse. This detrimental effect can be avoided if sufficiently strong mode-coupling mechanism is used. The methods used to lock the modes can be divided into two broad categories: active and passive. Active mode-locking is realised by introducing into the laser cavity an electro- or acousto-optical modulator driven with a frequency equal to the frequency spacing of laser cavity modes. The modulation of either amplitude or phase imposed on the beam inside the cavity creates side-bands for each mode. Because the modulation frequency is chosen for the side-bands to match the adjacent modes' positions the modulation provides coupling between modes. Active mode-locking is quite complex; one has to deal with RF signals and the modulation frequency has to match mode spacing with high accuracy. The main limitation of this method, however, is that it cannot be used to generate femtosecond

pulses. In the passive mode-locking no externally driven intra-cavity modulators are used. Instead, a suitable nonlinear component is introduced into the cavity. The role of this component is to modulate losses and/or gain of the laser so high instantaneous intensities are favoured. Given such a mechanism the intra-cavity light intensity will evolve in such a way that stronger fluctuations will grow at the expense of the smaller ones. Eventually only one such fluctuation will survive – one short pulse circulating inside the cavity. The nonlinear effects that can be used to achieve that are: saturable absorption, saturable gain, self-phase modulation and self-focusing. When discussing passive mode-locking it is more convenient to use a time-domain picture. In such a picture the response time of the nonlinear materials has to be taken into consideration. To achieve extremely short pulses one has to use sufficiently fast nonlinear effects. This can be achieved for a proper combination of saturable absorber/saturable gain [7] (dye lasers), with Kerr lens effect [4] (solid state lasers) or self-phase modulation [8] (fibre lasers). The Kerr lens mode-locking mechanism, now commonly used in Ti:Sapphire and other lasers based on vibronic gain media, can be explained as follows. The index of refraction of any material is constant at a given wavelength only in the regime of low light intensities. For higher intensities, however, the refractive index scales linearly (to the first approximation) with the light intensity $n(I) = n_0 + n_2 I$.

Let us now consider a gaussian beam inside the laser cavity. The very same Ti:Sapphire rod (typically of a length of several millimeters) that acts as a gain medium will behave like a focusing lens for sufficiently high light intensities. Furthermore, the focal length of such lens (the Kerr lens) decreases with increasing intensity (see Fig. 4). By combining this effect with an appropriate aperture placed inside the laser cavity, an intensity-dependent loss mechanism may be introduced. The Kerr nonlinearity is a non-resonant effect with response time below 1 fs and as such can be used for generation of very short pulses. As for the latest incarnation of the saturable absorber technique, multilayer dielectric mirrors with GaAs/AlGaAs quantum wells nicknamed SESAM (SEmiconductor Saturable Absorption Mirror) [9] are used. The reflection coefficient of such structure increases with increasing incident light intensity. With the record short pulses from Ti:Sapphire laser a combination of Kerr lens effect and saturable absorption mirror lead to impressive results [1]. The mode-locking is achieved in somehow different way in fibre lasers. Here, as the beam's transversal dimensions are determined by the mode diameter of the fibre, one cannot take advantage of the Kerr focusing, and nonlinear polarisation effect or self-phase modulation are used instead [10].

The main obstacle in generating very short laser pulses comes from the dispersion effects. The effect of the cavity dispersion can be easily understood in the frequency domain with the help of Eq. (1). Dispersive cavity means that the mode spacing is no longer a constant, the sum in Eq. (1) is different, and the resulting laser pulses are longer. Alternatively, one can analyse the problem in terms of a short pulse circulating inside the laser cavity. With proper design the pulse experiences shortening due to nonlinearity built into the cavity, ideally, until it reaches Fourier transform limit defined by the spectral width of the gain medium. However, at the same time the pulse will undergo broadening in time domain because of the group velocity dispersion (GVD) of the cavity. Typically, the second effect sets a limit on the pulse duration somewhere in the ps range unless the cavity is designed to minimise the dispersion. This can be done by incorporating a prismatic GVD line [11] and/or negative GVD mirrors [12] into the cavity. The prismatic dispersion line usually consists of two Brewster-cut prisms both used at the minimum deviation angle (Fig. 5). The first prism introduces an angular spread of different spectral components present in the laser beam. The role of the other prism is to reshape the beam, so that all the wavelengths propagate in parallel after it. This way a collimated beam leaves the prismatic line with different spectral components shifted in time with respect to one another due to different optical paths they had to travel. This shift (delay) for a given wavelength depends on the dispersion properties of the prisms material and the set-up geometry (prism separation and light path length in each prism). In a nega-

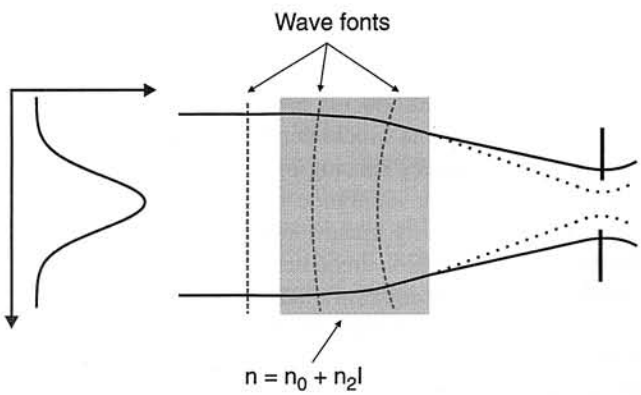

Fig. 4. Kerr lens with a slit. The dotted line corresponds to higher intensity. Due to changes in the focal length of the induced lens the loss of the transmitted beam decreases with increasing intensity.

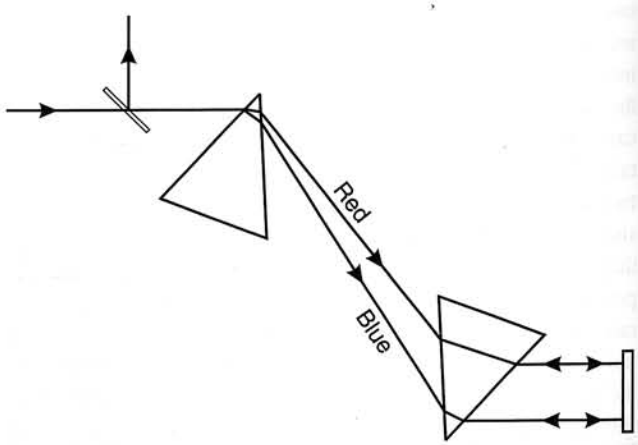

Fig. 5. Negative group velocity dispersion line built of two prisms.

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tive dispersion mirror (commonly referred to as a "chirped mirror") [13], the number and thickness of dielectric stack layers are designed in such a way that different wavelengths penetrate different depths into the mirror before being reflected therein. Thus, again, the optical path varies with wavelength in a prescribed way. As there are many more degrees of freedom (number of layers, thickness of each layer and its index of refraction) the dispersion of the laser cavity can be shaped accurately and compensated within an enormous bandwidth of hundreds of nanometers allowing generation of the shortest pulses. A different approach to overcome the dispersion effects is found in fibre lasers generating short pulses. Here, a number of fibre stretches with opposite signs of dispersion within the pulse spectrum are used to obtain zero total dispersion. One should also realise that the dispersion appears at all stages of ultrashort pulse handling. A fourier-limited gaussian pulse of 30 fs duration FWHM will be stretched to 37 fs after propagating through a 5 mm thick plate made of a typical optical glass (BK 7). Even worse temporal as well as spatial distortions of pulse front with respect to the phase front may appear during pulse focusing [14].

When applied outside the laser cavity the dispersion control may be used not only to compress ultrashort pulses to their minimum (bandwidth-limited) duration – it also allows shaping them to a desired form. As there is a need for such tailored pulses for example in the quantum control of chemical reactions with light, a number of techniques have been developed to actively control the dispersion. One of the techniques requires a multi-element phase (and sometimes amplitude) modulator, for example one based on liquid crystals [15]. If such a modulator is placed in the focal plane of the, so-called, 4f zero-dispersion arrangement (see Fig. 6) it can add a desired phase shift to each spectral component of the pulse spectrum which leads to a prescribed pulse shape. Similar methods involving deformable mirrors were also proposed and demonstrated [16]. Another approach to pulse shaping is to affect the pulse without prior spectral decomposition. A programmed pattern of a sound wave co-propagating with the pulse in a birefringent crystal may switch different spectral components from one polarisation to another at certain points along the pulse propagation direction. Since the two polarisations travel with different speeds within the crystal each spectral component experiences different programmable phase delay resulting in the pulse envelope shaping [17].

The selection of good gain media for femtosecond oscillators is limited and ultra-short light pulses can be obtained directly from lasers only in the visible and near infrared regions. Therefore it is of great importance that methods which enable fs pulse generation in other spectral regions are available. Fortunately, the last decade brought new and exciting developments in this area. However, many of the methods for femtosecond frequency conversion require pulse energies that are not available directly from oscillators and some amplification is required. Therefore, before describing frequency conversion techniques for femtosecond pulses, we will briefly discuss fs pulse amplification.

A femtosecond pulse laser amplifier differs from a standard long-pulse laser amplifier in two respects. First, as it has been discussed, propagation through the amplifying dispersive medium leads to temporal pulse broadening. Second, even with moderate pulse energies, the instantaneous power can be excessively high. It is easy to compensate the GVD effects in the amplifier by adding an external negative GVD line based on prisms, diffraction gratings, mirrors or a mixture of those components. Programmable pulse shapers discussed above can be also used for this purpose. However, high instantaneous power can cause unacceptable pulse distortions because of the self-induced phase modulation. Even worse, for high enough powers dielectric break-down in the amplifying medium leads to its irreversible damage. A standard way to avoid these problems relies on using the method called chirped pulse amplification (CPA) [18]. In this scheme an ultra-short laser pulse to be

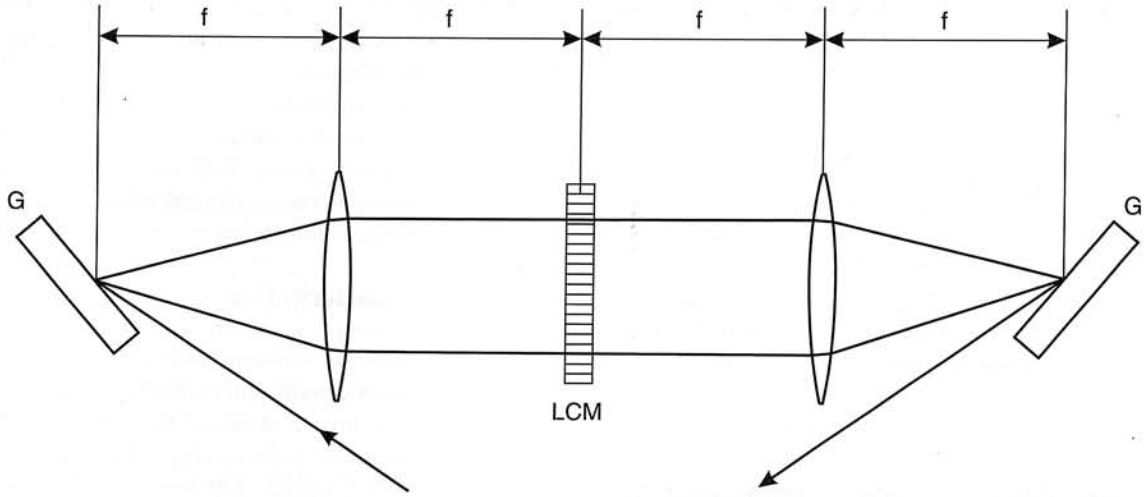

Fig. 6. Phase shaper based on 4f zero dispersion arrangement and a liquid crystal modulator (LCM). G – diffraction gratings, f – the lenses' focal length.

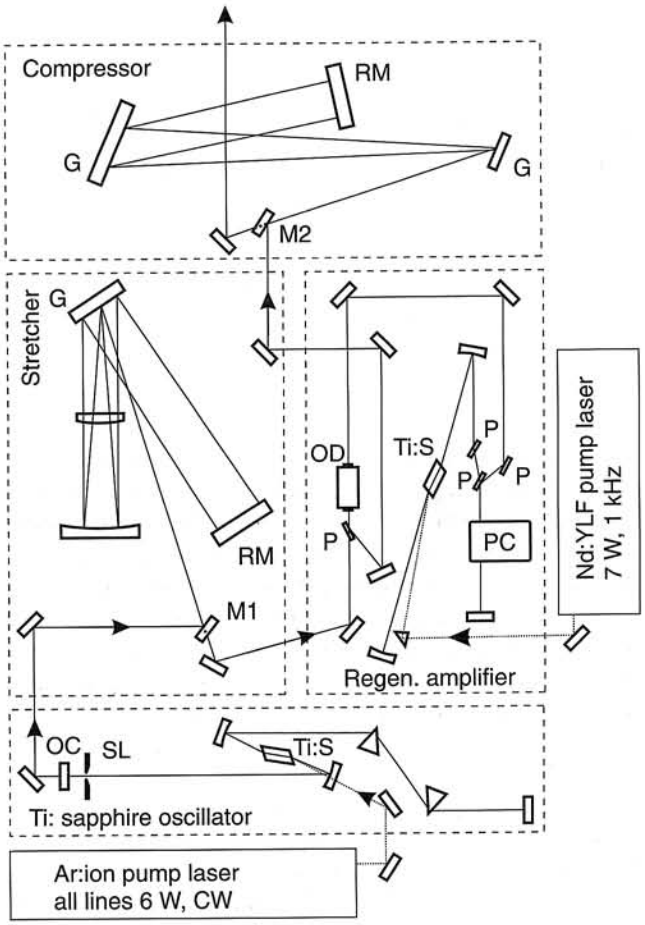

Fig. 7. CPA amplifier system. Ti:S – titanium-doped sapphire crystal, SL – adjustable slit, OC – 12% output coupler, G – 1200 grooves/mm diffraction gratings, RM – roof mirrors arrangements, P – polarising beam splitters, OD – optical diode, PC – Pockels cell; mirrors M1 and M2 are placed above the beam plane; all steering mirrors are broadband dielectric.

amplified is first stretched in time, amplified and finally compressed to near its initial duration. A typical commercially available CPA laser amplifier is shown in Fig. 7. The pulses of 30 nm spectral width (FWHM), 30 fs temporal width (FWHM) and 5 nJ energy from a Kerr lens mode-locked Ti:Sapphire oscillator are first injected into a grating stretcher (with a 1200 grooves/mm grating) in, for example, the Offner triplet [19] configuration. The pulses are stretched to a few hundred of ps duration and amplified some 10^6 times in the regenerative amplifier based on the Ti:Sapphire crystal pumped with 7 W of power from frequency-doubled Q-switched Nd:YLF laser operating at 1 kHz repetition rate. The pulse to be amplified is let into the amplifier cavity at a precisely chosen moment with respect to pump pulse that creates population inversion in the amplifying medium. After extracting the energy from the amplifier crystal, the pulse is ejected from the cavity. Pulse injection and extraction is performed with polarisation rotation introduced by a fast Pockels cell and a suitable set of thin film polarisers. The compression of amplified pulses is achieved with a compressor consisting of two 1200 grooves/mm gratings. An alternative for the regenerative amplifier is a multipass one [20] in which the number of pulse round-trips in the amplifier cavity is set with appropriate geometry rather than with the Pockels cell timing. With the techniques mentioned above even compact laboratory laser systems can reach very impressive powers in the range of tens of TW.

With the pulses of high enough energy one can use standard techniques for frequency conversion based on nonlinear processes. UV femtosecond pulses can be obtained by higher harmonics generation and frequency mixing. However, harmonics generation with ultra-short pulses poses specific challenges. Because the pulse spectrum is so broad special care has to taken to avoid spectral filtering resulting from the finite bandwidth of the process due to phase matching. Spectral filtering puts a severe limitation on the crystal thickness which, in turn, limits the efficiency of harmonics generation. As an example consider type I (*ooe*) second harmonic process in the commonly used BBO (β-barium borate) crystal. In this case, in order to support an 800 nm-centred bandwidth of 17 nm, the crystal thickness may not exceed 0.3 mm. In addition, group velocity dispersion causes that pulses with different frequencies propagate inside the nonlinear crystal with different speeds. In case of the third harmonic generation with two input pulses (fundamental and second harmonic) group velocity difference between those pulses limits the interaction length and efficiency. Both phase matching and group velocity mismatch cause that the harmonics pulse is usually longer then the fundamental frequency pulse. Group velocity mismatch can be eliminated when non-collinear geometry is used [21].

To have a continuously tunable source of femtosecond pulses one can use either optical parametric oscillator (OPO) or optical parametric generator (OPG). The second device is especially handy since it does not require a cavity. Using the available nonlinear crystals allows one to generate ultra-short pulses anywhere from visible to mid-infrared region [22,23].

Yet another source of tunable fs pulses is the process called supercontinuum generation discovered in the 1980-ties [24]. When an intense fs pulse is focused in a dielectric medium a mixture of 3rd order nonlinear processes leads to generation of a radiation pulse with a very broad spectrum (hundreds of nm). With an appropriate filter one can select a band of frequencies out of this continuum and thus obtain an ultra-short pulse at the desired wavelength.

3. Pulse characterisation

One can use standard techniques to measure many parameters of femtosecond light pulses. For example, with a low repetition rate source a standard laser energy meter will suffice to evaluate the pulse energy. Given the broad spectrum of ultra-short pulses it is not difficult to determine both the carrier frequency and the shape of the spectrum with the help of even a low-resolution spectrograph. The

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most difficult to measure and, at the same time, the most interesting is the temporal profile of the pulse. In order to fully characterise the pulse one has to determine its time-dependent electric field $E(t)$ which is typically written in the complex representation as

$$E(t) = A(t)e^{i(\omega_0 t+\varphi(t))} \tag{2}$$

where $A(t)$ is the slowly-varying pulse envelope, ω_0 is the central frequency, and $\varphi(t)$ is the time-dependent phase. However, in many cases one does not need all the information contained in $E(t)$. It is often sufficient to be able to estimate the pulse duration which can be defined as FWHM of the intensity profile $I(t)$. Unfortunately no electronic devices are fast enough to record pulses shorter than approx. 1 ps and indirect, optical methods have to be employed. The most popular method relies on experimental determination of intensity autocorrelation function A_c which is defined as

$$A_c(\tau) = \int_{-\infty}^{\infty} I(t)I(t-\tau)dt \tag{3}$$

where $I(t)$ is the pulse intensity and τ is the time delay between two replicas of the pulse to be measured. To record intensity autocorrelation function one needs a second order process. Originally, second harmonic generation process has been used for this purpose. In this scheme two replicas of the pulse to be measured cross at a small angle inside a suitable nonlinear crystal and the resulting second harmonic radiation is spatially filtered and recorded (e.g. with a photomultiplier tube) versus the delay between the pulses. A much simpler scheme based on two-photon absorption in a photodiode has been proposed [25] and successfully used as well. The photodiode provides a wide bandwidth and allows for a design of a compact autocorrelator. One such recently proposed device based on a Mach-Zechnder arrangement [26] is presented in Fig. 8. A rotating glass plate provides a time-dependent delay between the pulse replicas. A small number of elements guarantees little pulse distortion within the autocorrelator. Further, both arms contain the same set of optical elements, so the two pulse replicas reaching the detector are identical. The Mach-Zechnder arrangement was chosen because of its insensitivity to the input beam direction and position. It is not possible to retrieve $I(t)$ from the experimentally obtained $A_c(\tau)$. However, if one assumes a specific shape for the intensity profile then from the width of the intensity autocorrelation function the width of $I(t)$ can be found. If the two replicas propagate collinearly in the crystal then one measures the so called interferometric intensity autocorrelation function

$$S(\tau) = \int \left\{ |E(t) + E(t+\tau)|^2 \right\}^2 dt. \tag{4}$$

This measurement provides more information then the standard intensity autocorrelation, which for a given pulse envelope lacks any dependence on the phase. An example

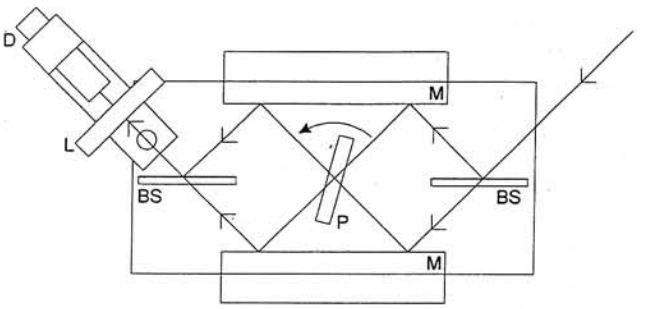

Fig. 8. A compact autocorrelator based on Mach-Zechnder configuration. BS – dielectric beam splitters, M – mirrors, P – rotating glass plate (1mm BK 7), L – focusing lens, D – light emitting diode.

of such measurement is presented in Fig. 9. It shows the interferometric autocorrelation function of 16 fs pulse from a home-built Ti:Sapphire laser recorded in our laboratory.

The autocorrelation measurements discussed so far require a train of pulses that are all assumed to be identical. It is also possible to collect the whole autocorrelation trace $A_c(\tau)$ using a single pulse. The scheme of such single-shot measurement is presented in Fig. 10. The two beams of pulse replicas are focused in the direction perpendicular to

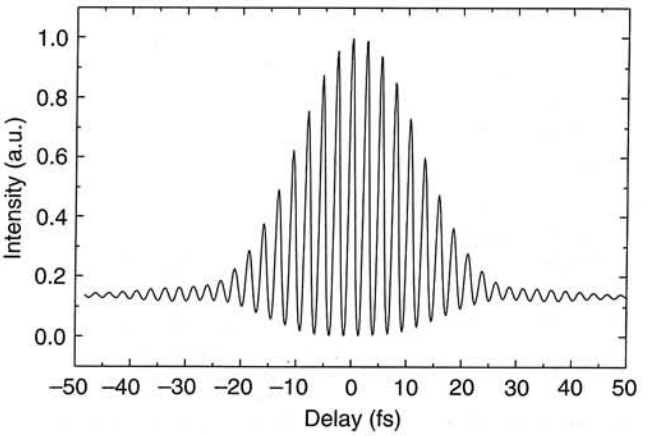

Fig. 9. Interferometric intensity autocorrelation of pulses from Ti:Sapphire laser.

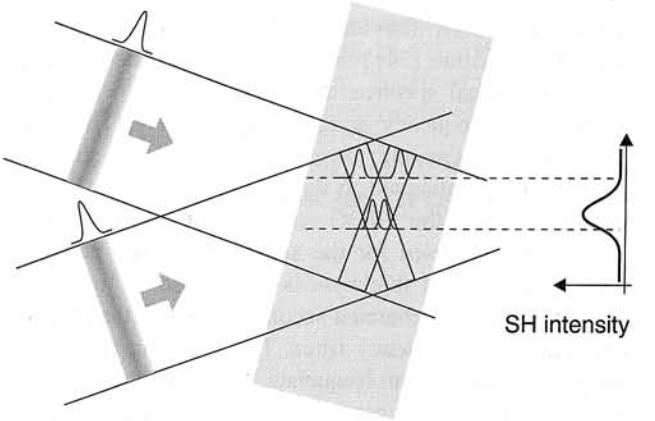

Fig. 10. Schematic of the single-shot measurement of the autocorrelation function. Delay between the pulses is a linear function of the position in the direction perpendicular to their propagation.

the plane of the drawing, while in the other direction they have finite transverse dimensions. One can easily see that the delay between pulses from both beams varies linearly with position. If the nonlinear crystal exit surface is imaged onto a one-dimensional detector (e.g. a CCD linear array) the autocorrelation resulting from a single pulse may be recorded.

The measurements get much more complicated when complete information on the pulse electric field is required. There has been much work done in this area in the past [27–31], yet out of many different methods proposed only two schemes are, at the moment, commonly used for complete amplitude and phase measurement of the pulse electric field. In a recently introduced spectral phase interferometry for direct electric-field reconstruction (SPIDER) method [32] an interferogram made of two frequency-shifted pulse replicas is recorded. To get these, first, three replicas of the pulse to be measured are formed and one of them is stretched with a grating stretcher or, for sufficiently short pulses, by propagation in a block of glass. Then, the two remaining time-shifted replicas are mixed with different spectral components of the stretched pulse in a nonlinear crystal. The resulting up-conversion signal is then recorded with a spectrograph and analysed. The method requires quite a complex optical set-up and relatively high-pulse energies. Its advantage lies in a non-iterative, unambiguous and fast (operating on a one-dimensional set of data) numerical procedure used to retrieve the spectral phase from the experimental data. FROG (frequency resolved optical gating) method [33] has been introduced about a decade ago and it has been extensively tested since. It is based on measuring spectrally resolved autocorrelation function – a spectrogram. Different nonlinear processes have been tested, among them second and third harmonic generation, polarisation gating and self-diffraction. Second harmonic generation, as a χ^2-based process, is the most sensitive but at the expense of the time direction ambiguity in the retrieved pulse. Some methods give intuitive traces that allow estimating pulse chirp at a glance, while other provide especially high bandwidth. As a rule, the best method is chosen individually for each measurement. An iterative algorithm [34] retrieves the electric field from the two-dimensional spectrogram by applying two constrains in time and frequency domains, respectively. The FROG apparatus is just an autocorrelator (CW or single-shot) with a spectrograph attached to its output. It has been recently pointed out that the FROG scheme can be simplified if a thick crystal is used for the second harmonic generation [35]. If the crystal thickness is chosen, so as to limit the bandwidth of the generated second harmonic, the crystal itself acts as a frequency filter. The resulting second harmonic (or rather a sum frequency, as two photons of different energies may take part in the process) is effectively generated within a certain spectral band. This band's width is inversely proportional to the crystal thickness and its central wavelength is defined by the crystal angle with respect to the beam's propagation direction. If the spatial distribution of the resulting sum-frequency light is recorded with a one-dimensional CCD array in the geometry shown in Fig. 10, than this is just one column (of constant wavelength) of the standard FROG spectrogram. The entire spectrogram is built of such records measured for different crystal angles. An example of the result obtained with this method is shown in Fig. 11. One can see that there is little distortion in pulse phase although the KDP crystal used here was 3-mm thick. This is the result of the trade-off between spectral filtering resolution and pulse distortion. For a short pulse that is easily affected by material dispersion the crystal thickness may be smaller as the spectrum is so broad it can be sampled with lower resolution. In case of longer pulses the spectral resolution, and thus the crystal thickness should increase but such pulses, having narrower bandwidth, are not so sensitive to propagation in dispersive media.

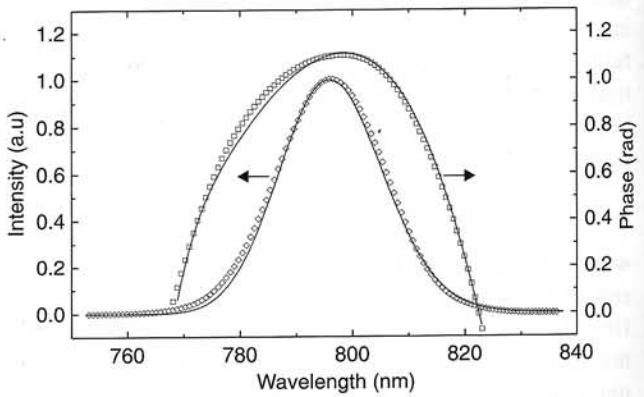

Fig. 11. Spectral amplitude and phase for pulses from a femtosecond Ti:Sapphire laser obtained with a standard FROG method (lines) and a thick crystal FROG method (diamonds and squares).

4. Prospects

What should we expect next? There is no doubt that the quest for shorter light pulses is far from over. It may seem that given the current state of laser technology the prospects are not that good because shorter pulses have to span even broader spectrum and finding optical components that can work in such a broad spectral range is not a simple task. On the other hand, very often, the progress results as much from the technological abilities as from new ideas that we cannot even try to foresee. As far as technology is concerned the main thrust will be probably towards simple, compact and reliable sources. Only such sources will make it possible to apply those pulses in industry, health care, etc.

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