

# Multiple ultra-short pulses generation for collinear pump-probe experiments

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A multiple pulses chirped pulse amplification (MPCPA) laser system was developed. The system can be used for multiple pulses generation in collinear pump probe experiments. In order to generate the multiple pulses, a simple passive pulse shaping technique in the spectral domain is implemented. The ratio of the intensity of the pulses and the delay between them can be controlled. Two and three spectrally separated pulses with duration of 300 fs and energies in the mJ range were generated, with intensity ratios from 0.05 to 1 and delays up to 600 ps.

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## 1. Introduction

Ultra-short laser pulses have a wide range of applications from triggering and tracing chemical reactions to important surgical applications in ophthalmology and neurosurgery, from testing ultrahigh-speed semiconductor devices to precision processing of materials (e.g. [1]), helping us to understand and control nature at very short time scales. In the optoelectronic domain, a range of nonlinear processes in transparent materials are actively studied (e.g. polymers [2], films [3]).

The Chirped Pulse Amplification (CPA) method, based on a temporal optical pulse stretcher and a compressor, was introduced in order to amplify ultra-short pulses to high intensities [4] needed in some experiments [5, 6]. Today, CPA technique is incorporated in most of the ultra-short and ultra-intense laser systems around the world [6].

While CPA method provides ultra-intense and ultra-short laser pulses, complementary studies are now oriented in the direction of "pulse-shaping" which allows control over amplitude, phase, frequency or inter-pulse separation [7]. Generation of two or more ultra-short pulses in a CPA amplification chain is of interest in experiments such as material processing [1], study of materials [2,3] and laser produced plasmas (e.g. x-ray lasers pumping [8]).

The usual pulse shaping methods and two or more pulses generation methods for pump-probe experiments are expensive or complicated to implement. Some are based on the design of a temporal pulse stretcher in a 4f configuration – for pulse envelope modulation [7] or on standalone interferometer set-ups (see e.g. [8]). In the present paper, we experimentally demonstrate an alternative, inexpensive method for the generation of multiple pulses [9,10], which is based on non-disturbing modification of an optical stretcher as found in most CPA laser systems. The difference resides in the fact that the modification of the amplitude or phase of the pulse is

made after the first pass of the initial short pulse through the stretcher, where the pulse is spatially stretched but still collimated, in contrast to the standard pulse shaping case where the spectral shaping is made in the focal plane of the focusing component of the stretcher [7]. As a consequence, with the method proposed here, one can delay parts of the spectrum and produce two pulses; this is not easily possible in the case of the traditional pulse shaping methods (such as 4f zero-dispersion line) [7] or in the cases based on interferometric splitting of the pulses [8]. The two pulses generated by our method are spectrally separated so they can be identified and monitored after their interaction with a given target, with the help of spectral dispersion methods. The method is also scalable, i.e. can be used to produce not only two but also three or more pulses.

The proposed method can be applied in a wide range of stretcher systems, including ones which are using prisms and gratings.

## 2. Principle of two collinear pulses generation in CPA systems

A typical CPA system can be modified, in order to produce two collinear pulses with complementary spectral components. A comparison of the standard CPA and multiple pulses CPA (MPCPA) architectures of the lasers is presented in the figure 1a. A laser oscillator produces ultra-short pulses with a broad spectrum. By introducing different optical paths for different spectral components, the pulses are stretched in time, thus reducing the peak intensity three to four orders of magnitude. Typically, the long wavelength spectral components ("red" part of the pulse) are coming earlier than the short wavelength spectral components ("blue" part of the pulse). The stretched pulses are amplified and recompressed to the initial short duration in the optical compressor. The

modification of the laser pulse at the stretcher level in the MPCPA architecture corresponds to the insertion of a temporal gap in the stretched pulse (figure 1b). In the MPCPA method described here, the duration of the gap and its position in the spectrum can be modified to match the experimental needs.

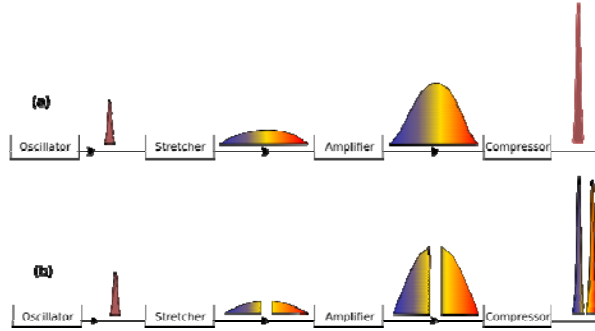


Fig. 1. Comparison between the standard CPA laser architecture (a) and multiple pulses CPA architecture proposed here (b).

Most of the pulse shaping techniques are related to the modification and control of the second and higher order terms of the Taylor series development (1) for the spectral phase of the ultra-short pulses  $\varphi(\omega)$ :

$$\varphi(\omega) = \varphi_0 + \frac{\varphi_1}{1!}(\omega - \omega_0) + \frac{\varphi_2}{2!}(\omega - \omega_0)^2 + \frac{\varphi_3}{3!}(\omega - \omega_0)^3 + \dots \quad (1)$$

where  $\omega$  is the electromagnetic field frequency and  $\omega_0$  is the central frequency of the spectrum of the laser pulse.

Our basic idea is to modify the **first order term**  $\varphi_1$  in the spectral phase distribution. This term corresponds to a simple temporal delay. In the stretcher, there is a proper place to do this, after the first pass through the stretcher, where one has a collimated beam with spatial chirp. The same approach is possible in the optical compressor after the first pass.

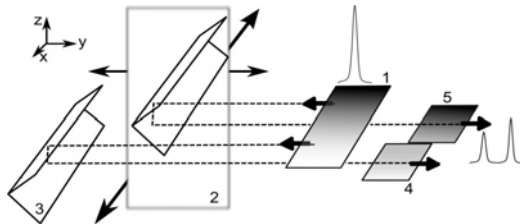


Fig. 2. Modification of the laser pulse after the first pass through the stretcher (or compressor) with the use of an additional, mobile roof mirror

In Fig. 2, we depict the principle of the multiple pulses generation used in our work. The laser system uses, at the stretcher or compressor level, a pulse laser 1, in the place where the spectral components of the laser pulse which propagate in the horizontal plane along the y axis

are spatially separated and collimated. This pulse first partially reaches the roof mirror 2 parallel to the axis x (for example the “blue” spectral components of the pulse), then the other part of the pulse reaches the roof mirror 3 parallel to the axis x, which is generally contained in the optical stretcher or compressor. A laser pulse 4, obtained by reflection on the roof mirror 3 has a longer optical path and will be delayed compared to the laser pulse 5 obtained by reflections on the roof mirror 2. After the final compression of the pulses 4 and 5 in the optical compressor of the laser system, two ultra-short laser pulses are obtained, that contain complementary spectral components. The control of the pulses delay between 4 and 5 is accomplished by changing the optical path difference between them by varying the distance between roof mirrors 2 and 3 along the y axis. The control of the pulses intensities ratio between 4 and 5 is accomplished by moving the roof mirror 2 parallel to the x axis and changing the contributing spectral domain of the pulses 4 and 5.

### 3. MPCPA set-up

We have built a CPA laser system using a commercial oscillator-regenerative amplifier laser system (Clark MXR-2101) as the femtosecond pulses source. The seed laser pulses have the central wavelength of 775 nm, about 8 nm spectral bandwidth FWHM, pulses duration of 190 femtoseconds, 4 mm diameter, running at 2 kHz with 0.7 mJ/pulse.

The numerical ray-tracing model, based on Rayica, a package for Mathematica, provides the wavelength-dependent phase for each ray of the optical stretcher and compressor systems, as described in [10].

Our optical stretcher, shown in the figure 3, includes a diffraction grating with 1200 lines/mm and 110x110x16 mm dimensions. The spherical mirror (SM) with 6 inch diameter has the focal length of 1200 mm and it is placed about 780 mm away from the diffraction grating. The vertical, rectangular flat mirrors M1 and M2 are used to build the all-reflective, folded, one-to-one telescope of the stretcher (M1 for folding the telescope, M2 in the focus). The fixed roof mirror (FRM) ensures the double passing of the optical system. After the stretcher, the laser pulse reaches 72 ps and the energy of the pulse is reduced to half. The stretched pulse is 20 times amplified in an 4-pass Ti:sapphire amplifier. The repetition rate of the amplifier is limited to 10 Hz.

The pulse reaches then the optical compressor, where it is compressed back to the original pulse duration but loses again half of the energy, reaching 3 mJ. In this CPA laser system we use only one diffraction grating for both stretcher and compressor as in ref. [11]. The compressor includes two roof mirrors to compensate for the optical path introduced by the stretcher. At the reference position  $D=0$  for the first compressor roof mirror, the compressor completely compensates for the optical paths differences introduced by the stretcher. The stretcher and compressor use the same incident angle on the diffraction grating.

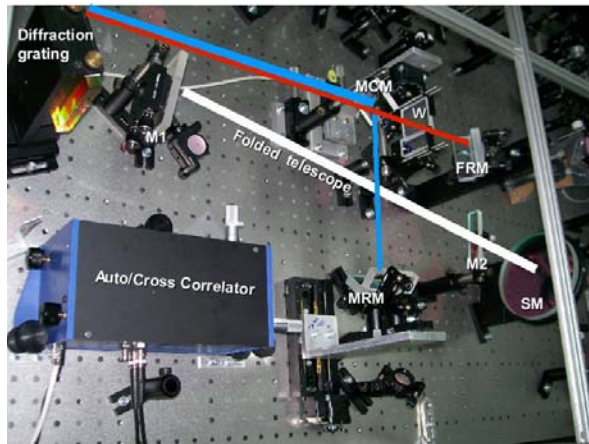


Fig. 3. Experimental set-up for multiple pulses generation in stretcher. The stretcher is formed by the diffraction grating, the folded telescope which uses the spherical mirror SM and two plane mirrors M1 and M2 and the fixed roof mirror FRM. The systems for multiple pulses generation is formed by the mobile clipping mirror MCM and the mobile roof mirror MRM or by the window W

For generating the two pulses in stretcher, we used an additional mobile clipping mirror (MCM), in order to deflect a part of the incoming laser pulse to the mobile roof mirror (MRM), as shown in figure 3. MCM was placed after the first pass of the laser pulse through the stretcher, before reaching the roof mirror FRM for a second pass, in the place where the pulse is spatially chirped and collimated. As a consequence, the blue parts of the spectrum are deflected towards the mobile roof mirror, while the red spectral components are passing by the mobile clipping mirror and reach the fixed roof mirror. The blue and red parts of the pulses have now different optical paths, depending on the position of the two roof mirrors relative to the mobile clipping mirror. After the second pass through the stretcher, the red and blue pulses are propagating collinear but with a temporal delay defined by the mobile roof mirror position. The pulses go through the amplifier and are compressed, in the end, in a standard single grating double pass compressor.

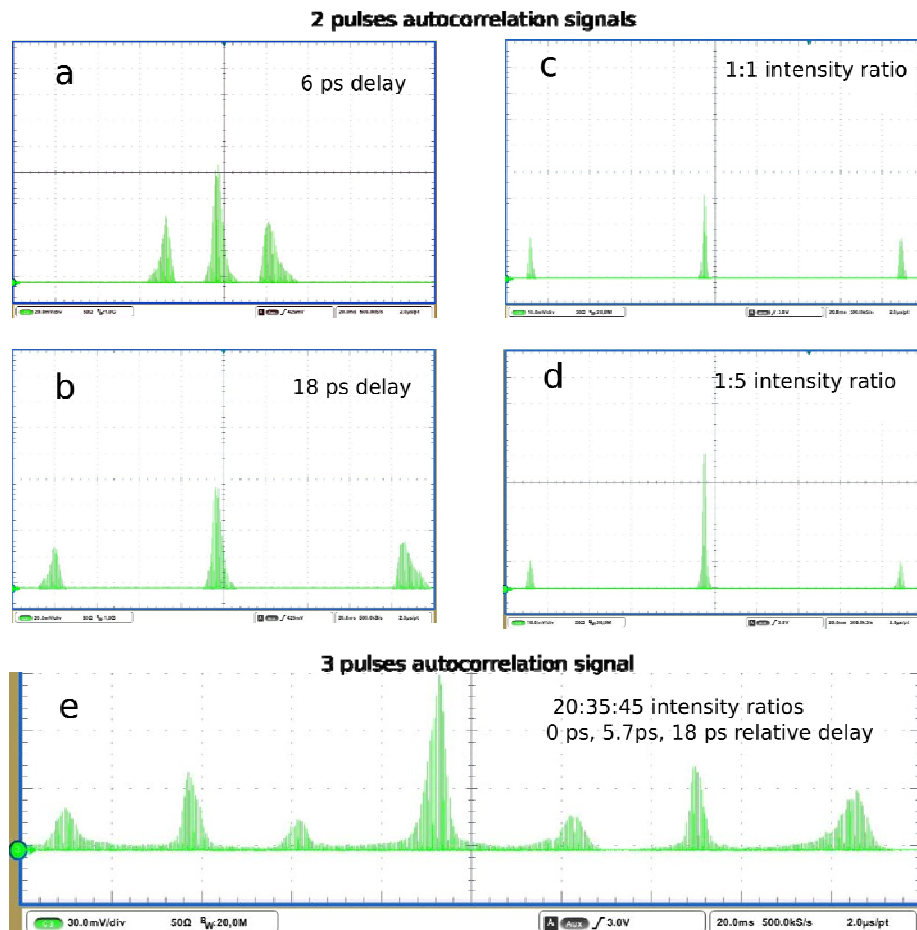
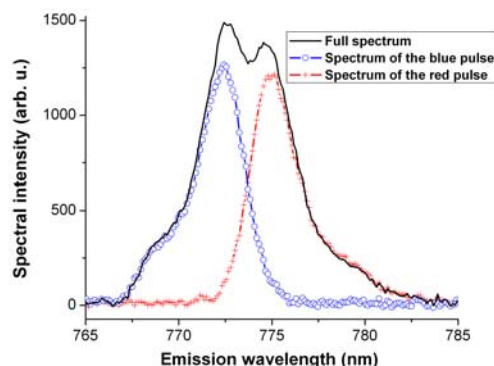


Fig. 4. Autocorrelation curves (20 ns/div temporal scale for all pictures) obtained with two pulses a) 1:1, 6ps delay, b) 1:1, 18 ps delay, c) 1:1, 18 ps delay, d) 1:5, 18 ps delay, and e) with three pulses with 20:35:45 intensity ratios and 0 ps, 5.7 ps and 18 ps temporal delays relative to a reference moment.

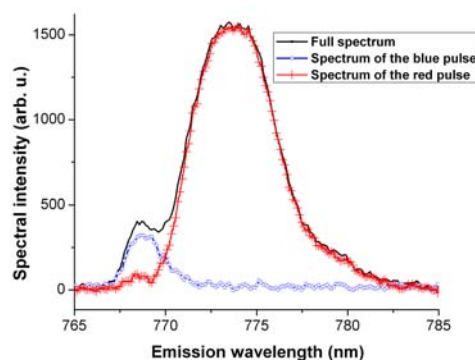
The identification of the two pulses was performed using a non-collinear second order autocorrelator from Femtochrome Inc (see fig. 3). The autocorrelation curves (obtained with 190fs/ms calibration factor) are presented in figure 4. It has a large temporal dynamics of up to 50 ps so we could identify the temporal structure we induced with our multiple pulse generation system (MPGS). As depicted in figure 4 a-d, the autocorrelation traces of the two pulses registered with a high speed Tektronix oscilloscope consists in three peaks. The central peak corresponds to the autocorrelation of the two pulses, while the first and the last signal correspond to the two cross correlations of the pulses. Shifting the mobile roof mirror from fig. 3, we could vary the delay between the pulses as depicted in figure 4a and 4b. In this figure the two pulses are equal in intensity, as the amplitude of the autocorrelation signal is double of the cross correlated signal amplitude. Maximum delay achievable with the system is 600 ps corresponding to 100 mm translation stage, with less than 5 femtoseconds temporal resolution. It was also easy to vary the pulse intensity ratio by translating the mobile clipping mirror MCM (fig. 3) in or out of the spatially chirped beam after the first pass in the stretcher.

We experimentally investigated also a complementary method to produce multiple pulses, by replacing the clipping mirror MCM and the mobile roof mirror MRM with an antireflection coated window W. In this case, the red and blue pulses are delayed due to the additional optical path introduced via the windows refractive index. This system can be used in the stretcher, as presented in figure 3, or it can be used in the compressor. In this case, the delay can be varied by inserting additional windows. The advantages of this approach are the simplicity (one optical element) and the fact that, by clipping the laser pulse, the delay between the red and blue pulses is not varied while the intensity ratio changes. We implemented a similar system in the compressor, too. We used a 10.5 mm BK7 plan parallel anti-reflexion coated window to obtain a delay of 18 ps, as registered with the autocorrelation system (figure 4c and 4d).

In order to prove the scalability of the system, we generated three pulses by combining the mobile roof mirror stretcher system with a windows based compressor MPGS. The autocorrelation trace allows identifying the intensity ratios and the delay of the pulses. The figure 4e shows the autocorrelation curve for three pulses with delays of 0 ps, 5.7 ps and 18 ps and with the intensity ratios of 20:35:45.



a



b

Figure 5: Spectral composition of two pulses generated in stretcher with 1:1 peak ratio (a) and 1:5 peak ratio (b)

The spectral composition of the pulses measured with an Ocean Optics HR4000+ spectrometer is presented in figure 5. The black continuous lines show the spectral composition of the two collinear pulses for different peak spectral intensity ratios, 1:1 (fig. 5a) and 1:5 (fig. 5b). The red curves (+) represent the spectra of the red pulses; the blue ones (o) show the spectra of the blue pulses, measured without the presence of the spectra of the red pulse. In both cases, there is a dip appearing in the total spectrum measured with both pulses at the same time, corresponding to the spectral position of the clipping. The spectral fingerprint of the MPGS is useful in the design and implementation of collinear pump probe experiments with ultra-short pulses. The remaining parts of the collinear pulses after the interaction with the target can be distinguished according to their spectral compositions.

#### 4. Conclusions

A simple method to generate collinear, spectrally complementary multiple pulses in a CPA laser system was presented. In CPA laser systems, the implementation of the method needs, as a minimum, only one optical component (the window) or two-three mirrors, while the stretcher and the compressor are already present in such CPA systems.

The delay and the intensity ratio of the generated pulses can be modified according to the experimental needs. The MPCPA method is scalable so the MPGS can be cascaded to produce three or more pulses. The system helps to investigate the materials dynamics under the action of ultra-short and intense laser fields with high temporal resolution.

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