# Generation of few-optical-cycle pulses tunable from the near to the far IR by optical parametric amplifiers.

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## Abstract

We exploit different optical parametric amplification schemes to generate ultra-broadband pulses with μJ-level energy broadly tunable from the near to the far-IR spectral region. In all cases we approach the single optical cycle limit with suitable compression techniques. Such pulses enable ultrafast spectroscopy on a variety of systems with unprecedented temporal resolution.

## 1. Introduction

Femtosecond optical pulses are powerful tools for investigating light-matter interaction, allowing the study of ultrafast dynamical processes in a wide range of materials. Pump-probe spectroscopy uses ultrashort pulses to track, in real time, the electronic and vibrational evolution of an optically excited sample. To be able to follow elementary photophysical and photochemical relaxation processes, it is necessary to push the temporal resolution, determined by the durations of pump and probe pulses, towards the limit set by the oscillation period of the carrier wave. In addition, the need to excite a system on resonance and probe optical transitions occurring at different photon energies requires broad frequency tunability of both pump and probe pulses.

Optical parametric amplifiers (OPAs) are known as flexible tools for the generation of broadly tunable ultrashort pulses. On the other hand, if suitably designed, OPAs act as broadband amplifiers, which can be used to dramatically shorten, by more than an order of magnitude, the duration of the driving pulse, down to the few-cycle regime [1]. In addition, OPAs can exploit non-linear optical interactions to achieve passive, all-optical CEP stabilization. In this presentation we review our research aimed at designing and implementing OPAs for the generation of broadly tunable few-optical-cycle pulses. We demonstrate different OPA schemes capable of producing 2÷3-cycle pulses with carrier wavelength almost continuously tunable from the visible to the mid-IR [2]. We also present the generation of high-field single-cycle multi-THz transients via OPA in a GaSe crystal. Peak fields exceeding 10 MV/cm and amplitude spectra covering more than three optical octaves are achieved. The temporal trace of the field is detected electro-optically with absolute phase and amplitude resolution.

## 2. Broadband Optical Parametric Amplification

To the first order, the gain bandwidth of an OPA can be expressed as $\Delta\nu \propto 1/|\delta_{si}|$ where $\delta_{si} = 1/v_{gs} - 1/v_{gi}$ is the group velocity mismatch (GVM) between the signal and idler pulses [1]. Therefore, when $v_{gs} = v_{gi}$ the bandwidth of the parametric amplification process becomes very broad. This condition can be achieved with three OPA configurations:

i) type I collinear interaction at degeneracy, for which the signal and idler pulses have the same polarization and frequency $\omega_s = \omega_i = \omega_p/2$;

ii) non-collinear interaction, for which the angle between signal and idler wave-vectors is chosen such that the signal group velocity equals the projection of the idler group velocity along the signal direction (non-collinear OPA, NOPA);

iii) generation of idler pulses in the spectral range beyond the zero dispersion wavelength (mid-IR OPA), in a collinear configuration.

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# 3 Few-optical-cycle pulse generation in near- and mid-IR

The three OPA schemes mentioned above all provide broad gain bandwidths; however, they also require suitable compression stages to manipulate the spectral phase and approach the transform-limited (TL) pulse duration. We implemented these three broadband OPA configurations using, according to the phase-matching configuration, either β-Barium Borate (BBO) or periodically poled stoichiometric Lithium Tantalate (PPSLT) as nonlinear media. The seed was provided by white light continuum (WLC) generated in a thin sapphire plate. Starting from a regeneratively amplified Ti:sapphire laser, we demonstrated the generation of ultrabroadband pulses covering the whole 1 - 4.5 μm spectral range.

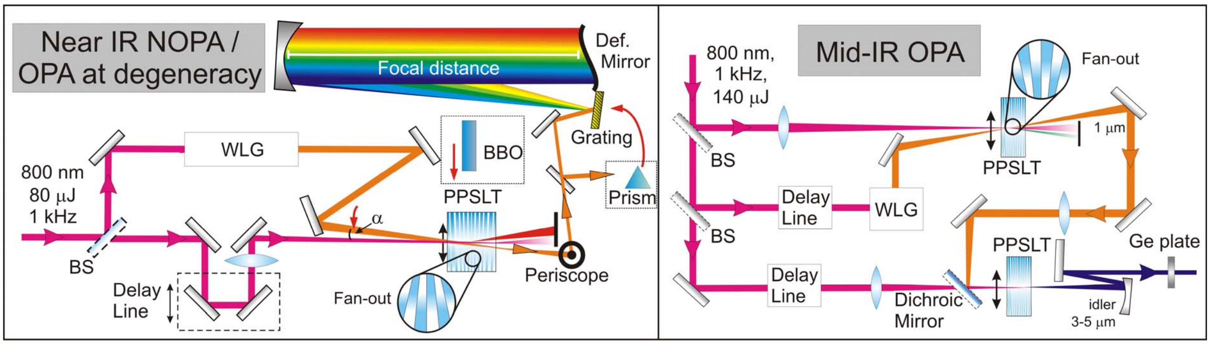

Figure 1. Schematic of the broadband IR OPAs: on the left the non-collinear geometry used to produce pulses in the 1000-1700 nm range, and the degenerate OPA producing broadband pulses at 1600 nm ; on the right the two-stage OPA generating pulses in mid-IR around 3500 nm. WLG: white light generation.

The near-IR degenerate OPA (Fig. 1, left panel) uses a 3-mm-thick BBO crystal cut for type I phase matching, pumped by 80 μJ of the Fundamental Frequency (FF) of Ti:sapphire; a small angle between pump and signal propagation direction allows separation between the spectrally overlapped signal and idler beams. The broadband amplified pulse, with a 1-2 μJ energy, extends from 1200 to 2200 nm (Fig. 1, dashed line) and supports a TL duration of less than two optical cycles. Fine control of the spectral phase is achieved by using a deformable mirror (DM) placed in the Fourier plane of a 4*f* shaper, equipped with a SF56 Brewster-cut prism as a dispersive element. At the optimal mirror deformation, we could generate pulses as short as 8.5 fs, corresponding to only 1.6 cycles of the 1600 nm carrier wavelength. The pulse temporal intensity, retrieved by Second Harmonic Generation-Frequency Resolved Optical Gating (SHG-FROG), is given in the central panel of Fig. 3.

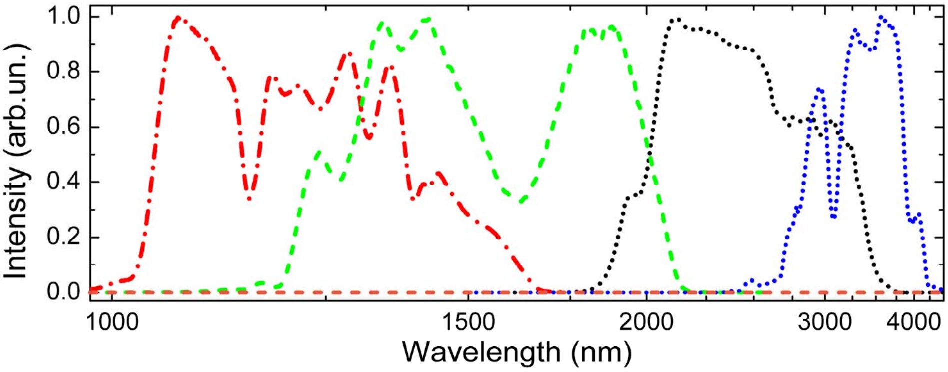

Figure 2. Series of spectra obtained by the three broadband OPAs, covering the 1000-4500 nm spectral range: near-infrared NOPA spectrum (dash dotted line); OPA at degeneracy (dashed line) and mid-IR OPA (dotted lines).

The second-harmonic pumped NOPA using a BBO crystal is a widely used source of few-optical-cycle pulses in the visible [3]. For this source the idler falls in the near-IR spectral range and has an higher group velocity with respect to the signal ($v_{gi}>v_{gs}$); this allows to match, in the non-collinear geometry, $v_{gs}$ with the projection of $v_{gi}$ along the signal direction. The same scheme can be exploited in the IR with FF pump; in this case, however, non-collinear group-velocity matching does not work with BBO, for which $v_{gi}<v_{gs}$, but requires high refractive index materials such as PPSLT [4], for which the zero dispersion wavelength is close to 2 μm.

A scheme of the near-IR NOPA is shown in Fig. 1, left panel. It employs a 1.2-mm-thick PPSLT crystal, designed with a fan-out (20.5- 23.7 µm) of the poling period to finely tune the amplification process and a non-collinear angle of 1.9°. We used 80 µJ of the FF to pump the NOPA and achieved amplified pulse energies up to 2 µJ. The amplified spectrum (dash-dotted line in Fig. 2) extends from 1000 to 1700 nm. Pulse compression was again performed with a DM system in a 4*f* shaper, coupled to a gold-coated diffraction grating as a dispersive element. The compressed pulse profile measured by SHG-FROG (Fig. 3, left panel) gives a pulsewidth of 8.5 fs, corresponding to about two cycles at 1300 nm [4].

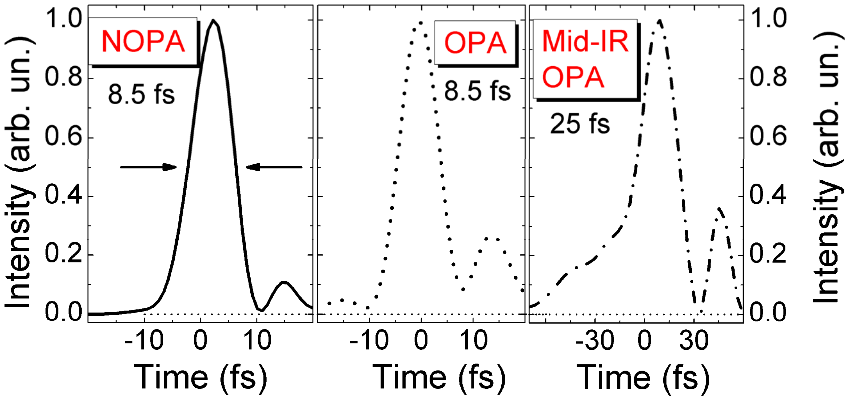

Figure 3: Pulse durations of the different IR OPAs retrieved by SHG-FROG measurements.

Group-velocity matched OPAs can be extended to the mid-IR range by exploiting the idler beam. High refractive index crystals such as PPSLT display, in a collinear geometry, another $\delta_{si}= 0$ point for signal wavelengths around 1 µm (and corresponding idler around 3-4 µm) [5]. While the supported gain bandwidth for the signal pulse is relatively limited (about 15 fs TL duration), it supports a sub-two-cycle pulse duration for the mid-IR idler wavelength. The setup for mid-IR pulse generation (Fig. 1, right panel) is based on a two-stage OPA using the same crystal as for the near-IR NOPA. The first stage is a slightly non-collinear WLC-seeded FF-pumped OPA, while the second one is strictly collinear, boosting the signal energy and generating the mid-IR idler free from any angular dispersion. Using only 120 µJ total pump energy (40 µJ for the first stage, 80 µJ for the second) we generated 2 µJ mid-IR pulses, tunable from 2 µm to 4.5 µm (dotted lines in Fig. 2). Pulse compression was obtained by bulk propagation in a 0.4 mm thick Ge plate, with a positive dispersion in the mid-IR compensating the second order negative dispersion accumulated during pulse generation and propagation in the PPSLT crystal. The pulse intensity profile, measured by SHG-FROG in a thin $AgGaS_2$ crystal, is reported in Fig. 2 and corresponds to a FWHM duration of 25 fs.

## 4. OPA for multi-THz pulse generation

Here we push the generation of ultra-broadband pulses to the multi-THz spectral range (≈15 µm). Our approach also exploits the generation of an inherently phase-stable idler pulse in a broadband OPA. To this end, we use a GaSe nonlinear crystal which displays an exceptionally large nonlinear coefficient. It allows for efficient phase-matching because of its strong birefringence. The flat dispersion slope of GaSe for long pump wavelengths leads to a relatively weak dependence of the internal phase-matching angle on the idler frequency. The broadest amplification bandwidth is expected for the case of perfect group velocity matching between signal and idler at an internal phase-matching angle of $\theta_{int}= 12.5°$. The THz spectrum is expected to show a broad peak at 24 THz and a side maximum at 50 THz. In contrast, THz generation via difference frequency mixing within the broad spectrum of a single femtosecond light pulse centered at 800 nm, i.e. optical rectification, suffers from substantial group velocity mismatch. To test the idea of pumping a multi-THz OPA with intense pulses at wavelengths longer than 800 nm, we use a hybrid laser system that combines the stability of Er:fiber technology with the high pulse energies of Ti:sapphire amplifiers. The regenerative Ti:sapphire amplifier is optically synchronized to the Er:fiber system and provides 5-mJ pulses at a repetition rate of 1 kHz. These pulses drive two identical two-stage OPAs which are seeded by the same white light continuum. One OPA is equipped with two BBO crystals, cut for type-I interaction, in order to generate broad signal spectra at a center wavelength of 1.28 µm. Pulse energies of 50 µJ and a temporal width of less than 30 fs are achieved after compression in a SF10-glass prism sequence. The second OPA is operated with two type-II-cut BBO crystals and provides pump pulses with narrower bandwidth centered at 1.18 µm with pulse energies of 200 µJ and a pulse duration of 115 fs.

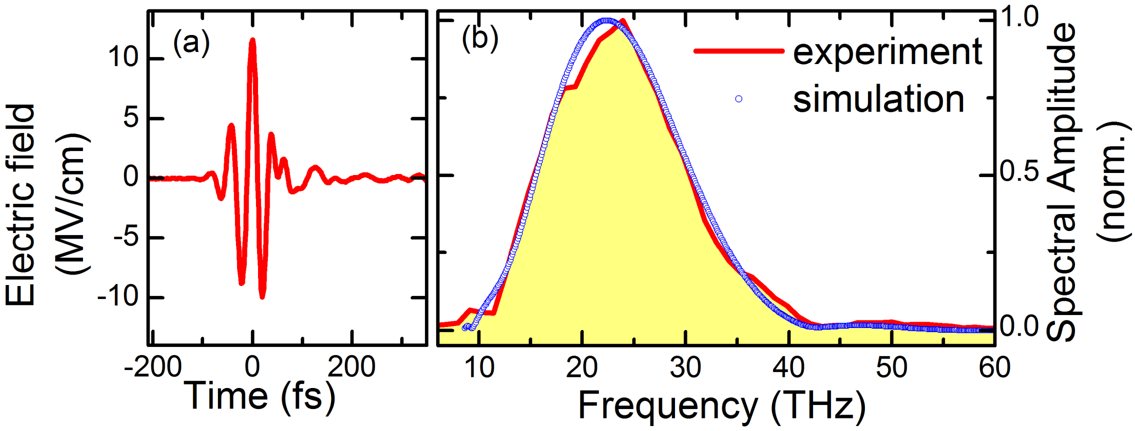

Figure 4: a) Multi-THz temporal profile as retrieved by electro-optical sampling; b) the spectrum covers the 10-40 THz window.

The signal and idler pulses are collinearly superimposed in the parametric mixing stage. We sample the temporal field trace of the emitted THz transients electro-optically by means of a 30-µm-thin GaSe detector and the near infrared 8-fs gating pulses from the fiber laser. The carrier-envelope offset frequency of the idler wave is expected to cancel during the OPA process due to the mutual phase coherence of the two OPA branches.

Figure 4(a) shows the electric field trace of the THz transient. It consists of only one pronounced peak in the time domain. The full width at half maximum (FWHM) of the intensity envelope amounts to 47 fs corresponding to 1.1 cycles at a center frequency of 24 THz (see the spectrum in Fig. 4(b)). With a calibrated thermopile detector, we measure an average THz power of 0.2 mW. This value translates into a peak electric field of 12 MV/cm. Those are by far the most intense single-cycle multi-THz transients known to us.

## 5. Conclusions

In this presentation we show that OPAs are able to generate few-optical-cycle pulses broadly tunable from the visible to the THz spectral range. Such pulses are expected to have a fundamental impact in ultrafast science and ultrafast spectroscopy where extreme temporal resolutions are needed.

## 6. References

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