

Millijoule few-cycle 5 μm source at 1 kHz repetition rate for generating broadband pulses from the mid- to far-infrared

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Abstract. We present a novel few-cycle 5 μm source delivering 75 fs pulses with 1.2 mJ energy at a 1 kHz repetition rate and its first applications for broadband pulse generation from the mid- to far-infrared.

So far, the lion's share of ultrafast spectroscopy is still performed with systems based on Ti:sapphire lasers. With such systems it is presently possible to generate ultrashort pulses covering a spectral range over seven orders of magnitude in frequency, i.e., from the THz to the x-ray range. However, at the edges of this spectral range, the conversion efficiency gets quite low. Recently, it has been found that pulses at longer wavelengths than Ti:sapphire, i.e., in the mid-infrared, have a much higher efficiency for the generation of ultrashort hard x-ray pulses [1]. On the other extreme, one expects a mid-infrared pump laser to be also more efficient in the generation of strong far-infrared pulses, needed, e.g., to study field-induced processes in condensed matter [2]. Thus, sources providing intense ultrashort pulses in the mid-infrared are of high current interest.

Here we present a novel high-power ultrashort pulse OPCPA source operating at kilohertz repetition rates with a center wavelength of 5 μm. This system provides 75 fs pulses with an energy of 1.2 mJ. As a proof of concept, THz pulses are generated by optical rectification in ZnTe. The setup used is sketched in Fig. 1.

The most promising route to access the wavelength range >4 μm together with scalability of the pulse energy relies on optical parametric chirped-pulse amplifier (OPCPA) using non-oxide crystals, like ZnGeP₂ (ZGP) or CdSiP₂ (CSP) [3]. For these nonlinear crystals a high-performance pulsed pump source with a wavelength ≥2 μm is required. The 2-μm Ho:YLF chirped pulse amplifier picosecond pump source delivering more than 50 mJ pulse energy at 1 kHz repetition rate with an excellent stability (pulse-to-pulse rms <0.3%) is described in [4]. A three color front-end based on a fs Er:fiber master oscillator operating at 40 MHz serves as seed for the 2-μm pump channel, the DFG produced at 3.4 μm and the gating pulse at 1.0 μm. The 3.4 μm pulses with a duration of 25 fs are used as signal for the optical parametric amplifier (OPA). The latter is composed

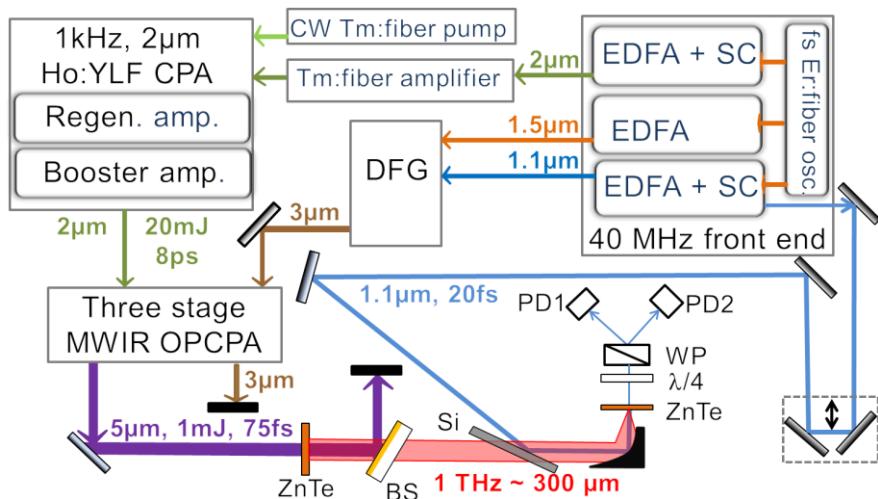


Fig. 1. Setup of the 5 μm driver source and THz generation and detection scheme. The main parts are the front-end, the difference frequency generation (DFG), the 2- μm Ho:YLF CPA amplifiers, the three OPA stages based on ZGP crystals and the THz generation and detection unit. EDFA, Er:fiber amplifier; SC, supercontinuum generation; Regen. Amp., regenerative amplifier; Booster amp., power amplifier; ZnTe, crystal for THz generation and detection; BS Broadband mid-infrared beam-splitter; WP, Wollaston prism; ZnTe, crystal for THz generation and detection; BS Broadband mid-infrared beam-splitter; WP, Wollaston prism; PD, photo diodes.

of three stages containing ZGP crystals as OPAs. Applying 14 mJ of the available 2 μm pulse energy in the third stage, 1.2 mJ per pulse is generated in the idler around 5 μm .

The 830 nm broad (FWHM) output spectrum of the idler (Fig. 2a) supports a Fourier-transform limited pulse duration of 60 fs. For the compression of the positively chirped idler pulses CaF₂ crystals are used. Compensating the residual group delay dispersion, the third- and fourth-order dispersion of the pulses by a spatial light modulator, successful recompression was achieved approaching the transform-limited duration, as determined by SHG-FROG measurements. The retrieved pulse shape is shown in Fig. 2(b). The pulse duration is 75 fs with an estimated energy content of 88%. The 75 fs pulse duration represents a record value for high energy mid-infrared OPCPAs, it corresponds to less than five optical cycles. The pulse energy of 1.2 mJ translates into a peak power of 14 GW, the highest for mid-IR OPCPAs beyond 4 μm so far. The emitted pulses are nearly diffraction-limited with a noticeable stability of the recompressed pulses with an rms of 1.3% [6].

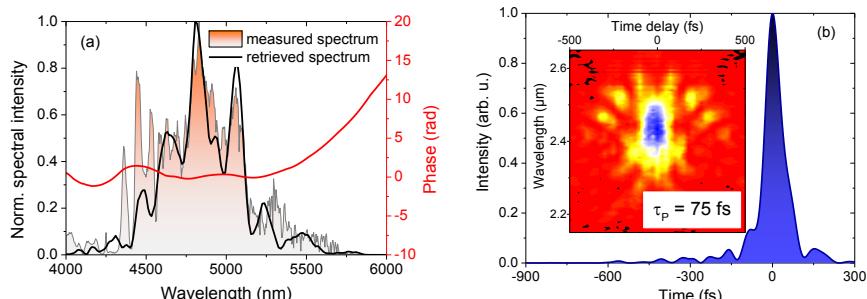


Fig. 2. SHG-FROG characterization of the 5 μm OPCPA pulses. (a) Optical spectrum, measured and retrieved (black). Phase (red); (b) retrieved temporal pulse shape (blue) and retrieved SHG-FROG trace (inset).

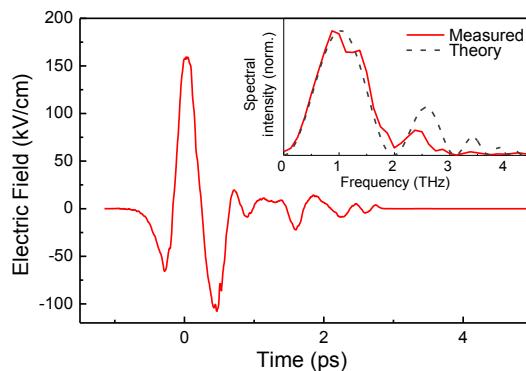


Fig. 3. THz transient generated in a 300 μm thick ZnTe crystal by optical rectification, and corresponding spectrum (inset): the dashed line corresponds to the expected output according to [5].

If focused to a 15 μm spot diameter, the pulses reach a peak intensity of $\sim 1.5 \times 10^{16} \text{ W/cm}^2$. The collimated 5 μm pulses can be used to generate far- and mid-infrared radiation by phase-matched difference frequency mixing in a nonlinear medium.

As a first example, the 5 μm pulses are used to generate far-infrared radiation by optical rectification in a [110] ZnTe crystal. The generated THz beam is separated from the pump by a broad-band mid-infrared beam splitter [7] and focused on a 300 μm thick ZnTe crystal, to be detected by electro-optic sampling [8]. As a probe, we use a 1.0 μm component of the supercontinuum, compressed to a pulse length of 20 fs. Figure 3 shows the generated THz electric field transient and the corresponding spectrum, centered at 1 THz. It is also possible to generate new wavelengths in the mid-infrared range by difference frequency mixing in organic crystals such as DSTMS [9], or by exploiting self-phase modulation of the 5 μm pulses in transparent dielectrics.

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