Multi-µJ 12 µm Femtosecond GaSe-based OPCPA at 1 kHz Repetition Rate

Pia Fuertjes, Martin Bock, Lorenz von Grafenstein, Uwe Griebner*, and Thomas Elsaesser

Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy, Max-Born-Str. 2a, Berlin, Germany D-12489
*griebner@mbi-berlin.de

Powerful ultrashort laser sources in the long-wave infrared (LWIR) spectral region are required for new applications in strong field physics, time-resolved vibrational spectroscopy of intra- and intermolecular low-frequency modes, surface plasmons, or strongly correlated systems. In particular, amplification systems in the wavelength range beyond 10 µm are limited by the availability and transparency of nonlinear crystals and there are only a few reports of femtosecond pulse generation exceeding 1 µJ energy [1,2]. Here we present a novel table-top LWIR OPCPA working at a 1 kHz repetition rate (ν_{rep}). It contains a femtosecond Cr:ZnS oscillator as front-end providing the seed for the 2-µm Ho:YLF pump and the signal at 2.4 µm without nonlinear conversion processes. In a 3-stage GaSe OPCPA the generation of sub-200 fs idler pulses at 11.4 µm with 50 µJ energy is demonstrated.

The setup of the fs Cr:ZnS laser front-end has been reported in ref. [3]. The emission spectrum of the Cr:ZnS laser (ν_{rep}=79 MHz), extending from 1.9 – 2.6 µm, is split at 2.1 µm by a dichroic mirror and the higher frequency part is used for seeding the pump. The 2-µm Ho:YLF regenerative amplifier pump delivers compressed pulses of a 3 ps duration and 12 mJ energy at ν_{rep}=1 kHz. The 46 fs signal pulses at 2.4 µm are stretched to 0.8 ps and subsequently phase-shaped by an acousto-optic programmable dispersive filter (Fastlite). For stretching the signal and compressing the idler pulses sapphire and ZnSe crystals are used, respectively.

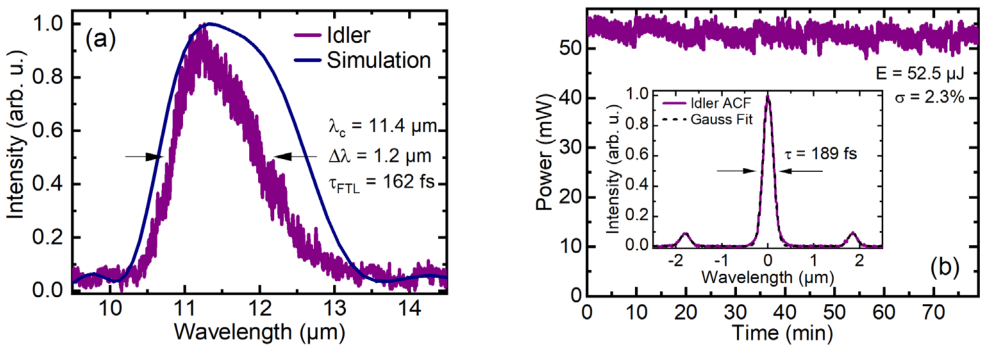

Fig. 1 Characterization of the idler pulses after the 3rd stage. (a) Measured and calculated spectrum (The latter is based on the signal spectrum after the 2nd stage.). (b) Long-term power stability; Inset: measured autocorrelation trace.

The LWIR OPA consists of 3 stages using GaSe as nonlinear crystals. In GaSe, the parametric process is phase-matched at an internal angle of 12.2° (Type-II). Employing overall 7 mJ of pump energy the combined signal and idler energy in the 1 kHz pulse train amounts to 310 µJ. The idler pulses are centered at 11.4 µm and contain 52.5 µJ energy. The resulting pump-to-idler energy conversion efficiency amounts to remarkable 0.75%. Figure 1a shows the idler spectrum after the third stage and the idler spectrum calculated with the measured signal spectrum after the second stage. A 1.2 µm bandwidth (FWHM) is achieved supporting a Fourier-transform limited (FTL) pulse duration of 162 fs. The bandwidth is limited by the phase-matching conditions for parametric frequency conversion in GaSe. Since the GaSe crystals are not anti-reflection coated, the high frequency modulation of the spectrum is associated with the 1 mm crystal thickness in the 2nd and 3rd stage. The measured autocorrelation trace of the re-compressed idler pulses is presented in the inset of Fig. 1b. Best fit to the data is achieved assuming a Gaussian-pulse shape and a duration of 189 fs is derived which corresponds to less than five optical cycles. The accompanying weak satellites are related to the modulations in the spectrum and carry about 5% of the energy. The estimated energy in the main peak amounts to 50 µJ which represents a record value for femtosecond laser sources beyond 10 µm [1,2]. The beam profile is nearly diffraction-limited with a remarkable stability of the recompressed pulses (rms: <2.5%). The latter is confirmed by a long-term power measurement shown in Fig. 1b.

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[2] R. Budriunas, K. Jurkus, M. Vengris, and A. Varanavicius, "Long seed, short pump: converting Yb-doped laser radiation to multi-µJ few-cycle pulses tunable through 2.5–15 µm," Opt. Express **30**, 13009 (2022).

[3] P. Fuertjes, L. von Grafenstein, D. Ueberschaer, C. Mei, U. Griebner, and T. Elsaesser, "Compact OPCPA system seeded by a Cr:ZnS laser for generating tunable femtosecond pulses in the MWIR," Opt. Lett. **46**, 1704 (2021).