Mid-IR short-pulse OPCPA with micro-Joule energy at 100 kHz

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Abstract: We present a novel mid-IR source based on optical parametric chirped pulse amplification (OPCPA) generating 96 fs pulses (9.0 cycles) at 3.2 μm with an energy of 1.2 μJ, at a repetition rate of 100 kHz. The amplified spectrum supports a minimum Fourier transform limited pulse duration of 45 fs, or 4.2 cycles. Our use of OPCPA allows the direct amplification of few-cycle pulses at this mid-IR wavelength, and is inherently scalable to higher energies. The seed source for the system is based on difference frequency generation (DFG) between two outputs of the same fibre laser: this source is expected to be intrinsically CEP stable.

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1. Introduction

Ultrashort pulsed light sources in the mid-IR are sought for numerous different fields ranging from fundamental strong field physics to medical and industrial applications. Strong field physics demands high repetition rate few-cycle pulses with stable carrier envelope phase (CEP). Generating such pulses in the mid-IR should result in shorter attosecond pulses [1, 2, 3], and allow improved discrimination between tunneling and multi-photon processes [4]. The broad bandwidths of such pulses are beneficial to spectroscopy since they cover many vibrational transitions in important molecules, and the intrinsic potential CEP stability adds a wide range

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of applications [5] such as breath monitoring for medical purposes [6]; the identification of bio-marker molecules [7]; monitoring the concentration of green house gases [8] or explosive detection via LIBS [9]. Here we present such a completely new, scalable and potentially CEP stable source based on OPCPA, generating 9 cycle pulses at 3.2 μm with 1.2 μJ energy at a repetition rate of 100 kHz.

Laser technology has advanced in recent years, enabling the production of near-IR pulse durations corresponding to only a few cycles of the laser field, at moderately high repetition rates and with stabilised CEP. These sources are nearly exclusively based on chirped pulse amplification (CPA) in Ti:Sapphire with subsequent broadening via gas-filled hollow fibres [10] or filamentation [11] and compression with chirped mirrors. State of the art pulse durations at centre wavelengths in the visible to near-IR currently lie in the few-cycle range at repetition rates up to a few kHz [12], but there exist significant motivations for the development of much less complex to operate new sources, particularly emitting at different wavelengths such as in the mid-IR (above 3 μm). A few-cycle mid-IR source has recently been demonstrated via four wave mixing in a filament [13], but this is based on a Ti:Sa CPA system, inheriting many of the associated disadvantages, and may not be scalable to high energies.

1.1. Mid-IR wavelengths

Mid-IR wavelengths are interesting, e.g. from an atomic physics application point of view, since they allow for a much clearer investigation of tunneling processes, whereas near-IR pulses operate in a mix of multi-photon and tunneling regimes. Attosecond pulses with carrier frequencies corresponding to extreme ultraviolet wavelengths can be produced from short-pulse laser systems, using high order harmonic generation (HHG) as coherent up-shifting mechanism from the near-IR drive laser [14, 15]. Here, changing drive laser wavelengths to the mid-IR is expected to yield shorter attosecond pulses due to a square of wavelength dependence of the shortest wavelength reachable via HHG [4, 16]. Recent experiments have confirmed this scaling of the harmonic cutoff with drive wavelength, while showing that predicted losses in harmonic yield [17] can be compensated by taking advantage of more favourable HHG phasematching at longer wavelengths [3]. Based on their results we expect that changing the wavelength of a typical NIR few-cycle source to 3 μm should allow generation of harmonic spectra extending to a photon energy well above 1 keV.

1.2. High repetition rates

Higher repetition rates help to improve signal to noise ratio for most experiments, but they are also essential for some in strong field physics; for instance, particle coincidence experiments with reaction microscopes (COLTRIMS) [18] permit the investigation of atomic and molecular processes with unprecedented scrutiny, but are limited mainly by the stability of current lasers due to the low cross sections of the processes under investigation; the measurement time is, in practice, nearly always longer than the time over which the best lasers can deliver constant performance. Using a 100 kHz repetition rate, experiments taking six days with a 1 kHz system can be completed in 90 min, greatly reducing the demands on the laser system stability.

1.3. CEP stability

Maintaining CEP stability is a key difficulty for most few-cycle laser sources, with current state of the art feedback stabilised systems capable of CEP locked operation for several hours, though locked durations of tens of minutes to a few hours are more common. Total CEP stability of the laser source is essential for many experiments, e.g. the measurement of double ionization [19], which demands a few-cycle laser source to be phase-stable with negligible amplitude instabilities and drift over about 12 hours with a 1 kHz repetition rate; an unrealistic requirement from

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current electronically CEP stabilised systems.

To date, various techniques allow the conversion of few-cycle near-IR sources to longer wavelengths, however these sources are based on Ti:Sa CPA, and inherit the associated problems of long term stability and operational complexity. DFG between different components of the same pulse followed by parametric amplification can create phase stable pulses with millijoule energies and few-cycle durations [20], but this is limited to near-IR and visible wavelengths. Parametric amplification of a white-light continuum can also provide short pulses in the visible-2 μm range [21], but as of yet has not been demonstrated to provide pulses in the mid-IR. The combination of Ti:Sa CPA and a laser diode in a frequency mixing scheme can produce short pulses in the mid-IR [22], but without CEP stability or easy scalability, with low efficiency and again requiring a complex CPA system as a front-end.

2. OPCPA layout

With the above-mentioned limitations in mind, we have designed a completely different mid-IR (3.2 μm) source with the goal for long-term stability and compactness based on optical parametric chirped pulse amplification (OPCPA) [23, 24]. The use of OPCPA as the amplification technique allows broadband amplification at arbitrary wavelengths [25], and does not use any gain storage as in a traditional laser system, thereby freeing the repetition rate of the laser from problems associated with thermal deposition in the amplifiers. Figure 1 shows the layout of this source, which incorporates the stability and ?hands-off? operability of a fibre front-end, stability of a diode-based pump laser, and the use of an intrinsically optically self-CEP-stable seed [26]. The source generates 96 fs pulses at 3.2 μm (9.0 cycles) with an amplified spectrum supporting a Fourier transform limited pulse duration of 45 fs, or 4.2 cycles. Currently we measure compressed pulse energies of 1.2 μJ, at a repetition rate of 100 kHz, however our use of OPCPA ensures this is inherently scalable to higher energies [27], with multi-Joule near-IR OPCPA sources already demonstrated [28].

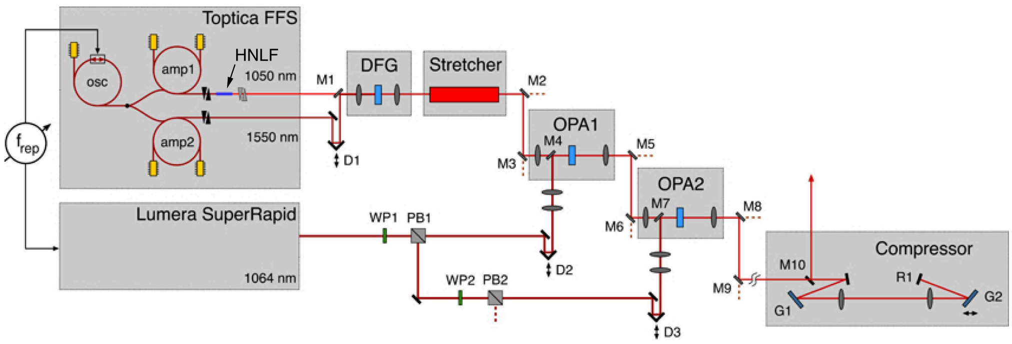

Fig. 1. Mid-IR OPCPA source layout. Two arms of a fibre laser (HNLF = highly nonlinear fibre) are combined through DFG to generate 3.2 μm radiation. This is then amplified by a double stage OPCPA pumped by a $Nd:YVO_4$ laser and finally compressed by a Martinez-type compressor. WP are waveplates, PB are polarising beamsplitters, D are delay stages, M1,4,7 are dichroic mirrors and other M represent gold mirrors.

The seed source for the system is based on difference frequency generation (DFG) between two outputs of the same fibre laser [29, 30]: this source is expected to be intrinsically carrier-envelope phase stable [26] since the fibre arms have been measured to show excellent relative timing stability, equivalent to a CEP jitter of 90 mrad over 1 s [31, 32]. The transfer of carrier-envelope phase ϕ in a three-wave mixing interaction such as DFG can be described by $\phi_p - \phi_s = \phi_i - \pi/2$ [26], where the subscripts p,s,i refer to the pump, signal and idler waves, listed in

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order of decreasing frequency. CEP fluctuations arise mainly in the oscillator, so that pump and signal waves produced from the same oscillator will have a constant phase difference $\phi_p - \phi_s = const$, creating an idler with fixed phase for use as our seed source. The phase stability of DFG generated pulses has been experimentally confirmed e.g. in Ref. [33].

OPCPA is also in principle perfectly CEP preserving, though in practice small phase matching errors between pump, signal and idler waves lead to some coupling between the pump intensity and CEP of the amplified pulse [34]. However, the additional phase noise induced is below the stabilised value for many conventionally amplified few-cycle systems, and so OPCPA amplification can be used to produce effectively CEP-stable, energetic pulses from a CEP-stable seed.

3. Mid-IR seeder

The mid-IR seed beam is generated by single path DFG between a broadband beam centred at 1.07 μm and a beam at 1.58 μm (1.07 μm$-$1.58 μm$\rightarrow$3.31 μm), both of which are produced by a commercially available fibre laser (Toptica FFS) (Fig. 1). The fibre laser operates at 100 MHz repetition rate and has two simultaneous outputs originating from a single oscillator, one providing 75 fs pulses at 1.58 μm with 160 mW average power while the second arm propagates identical pulses through a highly nonlinear fibre to generate a supercontinuum, from which a 1.0-1.15 μm spectral band is selected and compressed, yielding 65 fs pulses with an average power of 12.6 mW.

To create the difference frequency, the 1.07 μm and 1.58 μm pulses from the fibre laser were combined in a 2 mm long, periodically poled, MgO-doped Lithium Niobate (PPLN) crystal with a poling period of 31 μm. The crystals are heated to 80°C to avoid water absorption and minimise photorefractive damage. The two beams were collinearly focused onto the crystal, achieving respectively an intensity of 0.3 GW/cm^2 and 3 GW/cm^2. The resulting 3.2 μm DFG pulses are slightly down-chirped and have an average power of 1.6 mW at 100 MHz, corresponding to a quantum efficiency of more than 40% for the conversion process.

The spectrum of the DFG pulse was measured using a Fourier-transform IR spectrometer (FTIR) and is shown in Fig. 2. The spectrum spans from 2.9-3.8 μm at the $1/e^2$ level, and corresponds to a transform-limited pulse duration of 33 fs or 3 cycles. The DFG output beam is collimated by a Calcium Fluoride (CaF_2) lens, after which the residual 1 μm and 1.55 μm beams are rejected by the use of dielectric filters on CaF_2 substrates. The down-chirped DFG output is then further negatively stretched via propagation through an uncoated 10 cm long Sapphire rod, producing a 6 ps stretched pulse with which we seed the OPCPA amplifier chain.

4. Amplification

We amplify the stretched mid-IR seed beam, which is the idler wave, in two collinear geometry OPCPA amplifiers. The stability and beam quality of the pump laser in any OPCPA system is of crucial importance, and here we use a commercially available Nd:YVO_4 mode-locked laser (Lumera), delivering 10 W at 100 kHz with a pulse duration of 8.7 ps, and an extremely good spatial profile with an M^2 of 1.1. The fibre laser oscillator is electronically synchronised to the second harmonic of the 50 MHz oscillator of the Lumera laser, using a commercial synchronisation system (Toptica FFS-Sync-PLL). The RMS timing jitter between the two laser pulse trains is typically sub-350 fs. Note that since the pump pulse duration is $\sim$2 ps longer than the seed pulse duration, a temporal jitter of less than 5% of the pump pulse duration is negligible and excellent temporal overlap is ensured.

In the first parametric amplification stage the mid-IR seed is focused into a PPLN crystal identical to the DFG crystal. The pump beam power was set to 2.6 W via a series of waveplates and polarisers, generating a focused intensity of 60 GW/cm^2 at the crystal, producing a gain of

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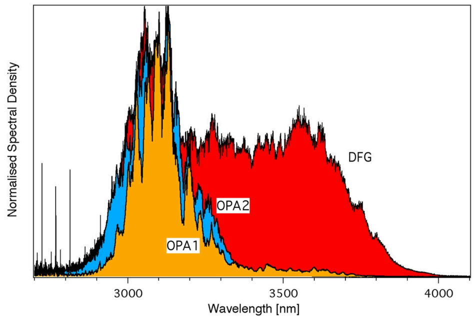

Fig. 2. Spectra at each nonlinear stage. The mid-IR spectrum after the difference frequency generation (red), first (orange) and second (blue) OPCPA stages.

4×10^3. The amplified mid-IR beam is recollimated and dielectric filters are used to reject the pump beam and any unwanted frequencies before propagation to the second OPCPA stage.

The second OPCPA stage again uses a 2 mm long PPLN crystal at the focus of the pump and seed beams, with identical characteristics to the DFG stage. The seed and pump beams are focused to intensities of 83 MW/cm^2 and 61 GW/cm^2 respectively. As in the first stage the beam is recollimated and passed through dielectric filters. The measured pulse energy after the two amplifier stages is 2.5 μJ at a repetition rate of 100 kHz. The amplified OPA1 spectrum is shown in Fig. 2, and it spans from 2.9-3.3 μm, at the $1/e^2$ level, corresponding to a transform-limited pulse duration of 45 fs, or 4.2 cycles at 3.2 μm. The loss of bandwidth on the long wavelength side of the spectrum is due to the phase-matching in the amplification process and affects only slightly the transform-limited pulse duration. The modulation seen on the spectrum has been identified as originating from etalon effects in the fibre laser which create double pulsing, and is not an artifact of the OPCPA process. Work is in progress to modify the cavity in order to suppress this effect.

5. Compression

To compress the down-chirped amplified pulse we used a Martinez type 4-f grating sequence with two 200 line/mm gold coated gratings designed for 3 μm. The total efficiency of the compressor is 48%, and our measured compressed energy is 1.2 μJ, with most losses due to the uncoated lenses in the setup.

The pulse duration is measured using second harmonic generation frequency resolved optical gating (SHG-FROG) together with the FTIR spectrometer giving a resolution of up to 0.5 cm^{-1}. The two replica pulses in the FROG are created using a pellicle beam splitter to minimise dispersion, and are focused non-collinearly into a 0.2 mm thick $AgGaS_2$ crystal whose acceptance bandwidth is over 1.8 μm at a central wavelength of 3.2 μm. To remove the time-reversal ambiguity in the FROG we directed the optimally compressed pulse through an 8 mm thick Silicon plate. Comparing the change in the FROG retrieved phase with the known phase introduced by the silicon allowed us to determine the correct direction for the time axis.

With our simple compressor, we have measured pulses as short as 96 fs, corresponding to 9.0 cycles at 3.2 μm, and within a factor of two of the 45 fs, 4.2 cycle transform limited pulse duration (Fig. 4). Note that we have highlighted the very small-scale features in the FROG trace (Fig. 3) over-proportionally through a square-root of the intensity colour scaling. As mentioned before, these features are known to arise from the fiber seeder and we have taken great care to record our FROG trace with high enough spectral resolution and over a very large time window, without which these subtleties are easily missed; see e.g. [30]. Recording such ultrahigh

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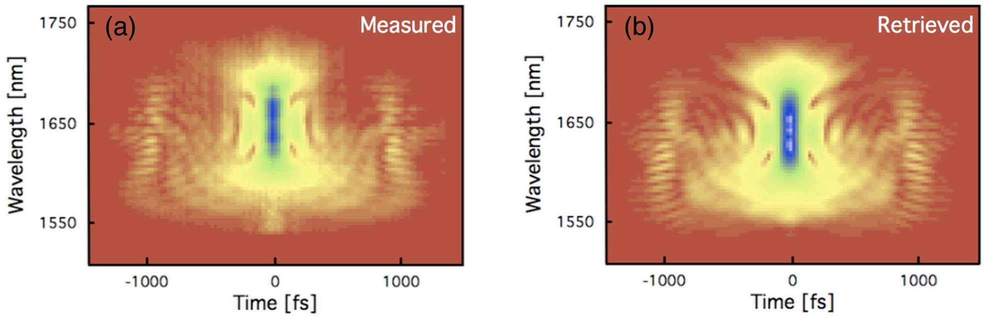

Fig. 3. Measured and retrieved FROG traces. (a) Measured FROG trace of the mid-IR pulse - the colour scale represents the square root of the normalised intensity, chosen to highlight the small features (b) Retrieved FROG trace of the mid-IR pulse, using the same colour scale.

resolution trace is not trivial in the mid-IR but the excellent agreement in the measured and reconstructed FROG traces demonstrate the high fidelity of our measurement and the fringes are of no big consequence for the temporal profile as can be seen from Fig. 4.

We also measured a knife-edge spatial profile of the compressed pulse 1.5 m after exiting the compressor (Fig. 4 inset), showing an excellent beam quality, with close to a Gaussian profile.

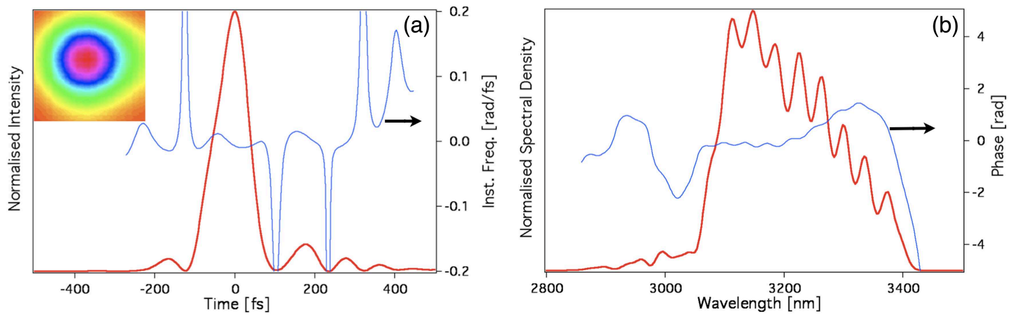

Fig. 4. Reconstructed pulse from the FROG measurement. (a) Temporal profile and instantaneous frequency retrieved by the FROG: 96 fs FWHM = 9.0 cycles (at 3.2 μm one cycle=10.7 fs) The spatial profile is shown in inset, width of image corresponds to 5 mm (b) Spectrum and phase retrieved by the FROG .

The energy achieved is sufficient for spectroscopy [5] as well as for some strong field physics experiments, since focusing to a few times the diffraction limit already yields intensities in excess of 10^{13}W/cm^2, and the total energy by no means represents the achievable limit from our system. As shot noise in experiments typically scales inversely with repetition rate, our 100 kHz system can produce data with higher signal to noise ratio than would be expected from comparable energies at kHz repetition rates. We expect our OPCPA design to be scalable in repetition rate and pulse energy with the limitation being the pump laser. Sources with average powers of up to 200 W are however already available and we can expect near linear scaling of the OPCPA output with the available pump laser pulse energy.

One issue that can limit energy scaling in OPCPA systems is parametric superfluorescence, although the level of fluorescence seen varies between systems [25, 35]. One successful approach to limit superfluorescence is the use of a high energy seed pulse [25], which however is

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inappropriate in our case since we have deliberately chosen to use a fibre laser to take advantage of its high stability at the expense of seed energy. In our system we maximise the pump-seed volume overlap in each OPA, choose closely matched pump and seed durations, and limit the OPA gain to less than 10^4 to help reduce the problem of unwanted superfluorescence, while the inclusion of dielectric spectral filtering between each OPA stage should further suppress fluorescence at unwanted frequencies.

6. Conclusion

To conclude, we have devised a completely new source of ultrashort laser pulses (9.0 cycles) in the mid-IR (3.2 μm) with a repetition rate of 100 kHz. This novel design is compact, solid-state, solely based on OPCPA, and incorporates a fibre seeder as well as solid-state pump laser, thereby ensuring maximum reliability and hands-off operation. The pulse energy, measured after compression, is 1.2 μJ and already sufficient for spectroscopy or even some strong field experiments. As shot noise in experiments typically scales inversely with repetition rate, our 100 kHz system can produce data with higher signal to noise ratio than would be expected from comparable energies at kHz repetition rates. The energy can easily be improved by replacing our metal optics (6% loss per reflection) with highly reflective all dielectric optics, or by upgrading our pump laser pulse energy.

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