

Mid-IR 6 cycle OCPA at 100 kHz**OPCPA de 6 ciclos en IR medio a 100 kHz**A. Thai^{(1,*),} O. Chalus^{(1),} P. K. Bates^{(1),} J. Biegert^(1,2)

1. ICFO-Institut de Ciències Fotoniques, Mediterranean Technology Park, 08860 Castelldefels (Barcelona), Spain

2. ICREA-Institució Catalana de Recerca i Estudis Avançats, 08010 Barcelona, Spain

(*) Email: alexandre.thai@icfo.es

Recibido / Received: 30/10/2010. Aceptado / Accepted: 15/12/2010

ABSTRACT:

We report on an optical parametric chirped pulse amplification source of ultrafast pulses in the mid-IR. The system is all solid-state, diode pumped, and operates at 100 kHz repetition rate. The 3.8 μJ energy pulses at 3.1 μm centre wavelength have record power stability of 0.75 % rms over 30 min and pulse duration of 67 fs or 6 optical cycles.

Keywords: Nonlinear Optics, Ultrafast Optics, OCPA.

RESUMEN:

Presentamos una fuente de pulsos ultracortos en el rango IR medio, basado en un sistema de amplificación óptica paramétrica. El sistema es totalmente de estado sólido, bombeado mediante diodo, y opera con un ritmo de repetición de 100 kHz. Los pulsos alcanzan una energía de 3.8 μJ en la longitud de onda central de 3.1 μm , con una estabilidad de 0.75% rms a lo largo de 30 minutos, con una duración de los pulsos de 67 fs o 6 ciclos ópticos.

Palabras clave: Óptica No lineal, Óptica Ultrarápida, OCPA.

REFERENCES AND LINKS

- [1]. P. Agostini, L. F. DiMauro, "The physics of attosecond light pulses", *Rep. Prog. Phys.* **67**, 813-855 (2004).
- [2]. P. B. Corkum, F. Krausz, "Attosecond science", *Nat. Phys.* **3**, 381-387 (2007).
- [3]. T. Popmintchev, M.-C. Chen, O. Cohen, M. E. Grisham, J. J. Rocca, M. M. Murnane, H. C. Kapteyn, "Extended phase matching of high harmonics driven by mid-infrared light", *Opt. Lett.* **33**, 2128-2130, (2008).
- [4]. B. Sheehy, J. D. D. Martin, L. F. Dimauuro, P. Agostini, K. J. Schafer, M. Gaarde, K. C. Kulander, "High harmonic generation at long wavelengths", *Phys. Rev. Lett.* **83**, 5270-5273 (1999).
- [5]. P. Mazzone, "Analysis of volatile organic compounds in the exhaled breath for the diagnosis of lung cancer", *J. Thorac. Oncol.* **3**, 774-780 (2008).
- [6]. K. Namjou, C. B. Roller, T. E. Reich, J. D. Jeffers, G. L. McMillen, P. J. McCann, M. A. Camp, "Determination of exhaled nitric oxide distributions in a diverse sample population using tunable diode laser absorption spectroscopy", *Appl. Phys. B* **85**, 427-435 (2006).
- [7]. F. Tittel, D. Richter, A. Fried "Mid-infrared laser applications in spectroscopy", *Solid-State Mid-Infrared Laser Sources* **89**, 445-510 (2003).
- [8]. D. Mirell, O. Chalus, K. Peterson, J.-C. Diels "Remote sensing of explosive using infrared and ultraviolet filaments", *J. Opt. Soc. Am. B* **25**, B108-B111 (2008).
- [9]. D. G. Lancaster, D. Richter, R. F. Curl, F. K. Tittel, L. Goldberg, J. Koplow, "High-power continuous wave mid-infrared radiation generated by difference frequency mixing of diode-laser-seeded fiber amplifiers and its application to dual-beam spectroscopy", *Opt. Lett.* **24**, 1744-1746 (1999).

- [10]. D. Faccio, A. Gruen, P. Bates, O. Chalus, J. Biegert "Optical amplification in the near-infrared in gas-filled hollow-core fibers", *Opt. Lett.* **34**, 2918-2920 (2009).
- [11]. T. Fuji, T. Suzuki, "Generation of sub-two-cycle mid-infrared pulses by four-wave mixing through filamentation in air", *Opt. Lett.* **32**, 3330-3332 (2007).
- [12]. V. Petrov, M. Ghotbi, O. Kokabee, A. Esteban-Martin, F. Noack, A. Gaydardzhev, I. Nikolov, P. Tzankov, I. Buchvarov, K. Miyata, A. Majchrowski, I. Kityk, F. Rotermund, E. Michalski, M. Ebrahim-Zadeh, "Femtosecond nonlinear frequency conversion based on BiB₃O₆", *Laser Photonics Rev.* **4**, 53-98 (2010).
- [13]. R. Kaindl, M. Wurm, K. Reimann, P. Hamm, A. Weiner, M. Woerner, "Generation, shaping and characterization of intense femtosecond pulses tunable from 3 to 20 μ m", *J. Opt. Soc. Am. B* **17**, 2086-2094 (2000).
- [14]. E. Nibbering, T. Elsaesser, "Ultrafast vibrational dynamics of hydrogen bonds in condensed phase", *Chem. Rev.* **104**, 1887-1914 (2004).
- [15]. M. Gertsvolf, H. Jean-Ruel, P. P. Rajeev, D. D. Klug, D. M. Rayner, P. B. Corkum, "Orientation-dependent multiphoton ionization in wide band gap crystals", *Phys. Rev. Lett.* **101**, 243001 (2008).
- [16]. F. Tavella, A. Willner, J. Rothhardt, S. Hädrich, E. Seise, S. Düsterer, T. Tschentscher, H. Schlarb, J. Feldhaus, J. Limpert, A. Tünnermann, J. Rossbach, "Fiber-amplifier pumped high average power few-cycle pulse non-collinear OPCPA", *Opt. Express*, **18**, 4689-4694 (2010).
- [17]. N. Ishii, L. Turi, V. S. Yakovlev, T. Fuji, F. Krausz, A. Baltuska, R. Butkus, G. Veitas, V. Smilgevicius, R. Danielius, A. Piskarskas, "Multimillijoule chirped parametric amplification offew-cycle pulses", *Opt. Lett.* **30**, 567-569 (2005).
- [18]. O. Chalus, P. K. Bates, M. Smolarski, J. Biegert, "Mid-ir short-pulse OPCPA with microjoule energy at 100 Khz". *Opt. Express* **17**, 3587-3594 (2009).
- [19]. C. Erny, K. Moutzouris, J. Biegert, D. Kühlke, F. Adler, A. Leitenstorfer, U. Keller, "Mid-infrared difference-frequency generation of ultrashort pulses tunable between 3.2 and 4.8 μ m from a compact fiber source", *Opt. Lett.* **32**, 1138-1140 (2007).
- [20]. C. Vozzi, G. Cirimi, C. Manzoni, E. Benedetti, F. Calegari, G. Sansone, S. Stagira, O. Svelto, S. De Silvestri, M. Nisoli, G. Cerullo, "High-energy, few-optical-cycle pulses at 1.5 μ m with passive carrier-envelope phase stabilization", *Opt. Express* **14**, 10109-10116 (2006).
- [21]. O. Chalus, P. K. Bates, J. Biegert, "Design and simulation of few-cycle optical parametric chirped pulse amplification at mid-ir wavelengths", *Opt. Express* **16**, 21297-21304 (2008).
- [22]. P. Bates, O. Chalus, J. Biegert, "Ultrashort pulse characterization in the mid-IR", *Opt. Lett.* **35**, 1377-1379 (2010).
- [23]. O. Chalus, A. Thai, P.K. Bates, J. Biegert "6 cycle mid-IR source with 3.8 μ m at 100 KHz", *Opt. Lett.* **35**, 3204-3206 (2010).

1. Introduction

The majority of recent results in the field of ultrafast physics have been driven by Ti:Sapphire lasers at 800 nm due to the stability, reliability and commercial availability of these sources. However, there are a great many applications that demand ultrafast pulses in the mid-IR spectral range, which are limited by the unavailability of appropriate sources. Within strong field physics, the generation of XUV radiation via HHG for imaging demands high repetition rate few-cycle pulses at longer wavelengths with stable carrier envelope phase (CEP). Intense mid-IR pulses could also lead to shorter attosecond pulse generation [1-3] and

clearer separation of multiphoton and tunneling processes [4]. The broad bandwidths of such pulses are beneficial to spectroscopy as well since they cover many vibrational transitions in important molecules opening a wide range of applications such as breath monitoring for medical purposes [5]; the identification of biomarker molecules [6]; but also monitoring the concentration of green house gases [7] or explosive detection via laser induced breakdown spectroscopy [8].

Current mid-IR short pulse sources are largely based on down-shifting of near-IR ultrashort pulses from amplified Ti:Sapphire chirped pulse amplification (CPA) systems. Mixing these pulses

with a second laser [9] can create mid-IR pulses, however this is unlikely to be practical for few-cycle pulse generation and does not produce CEP stability. Four-wave mixing of the pulses and their second harmonic in a filament or hollow fibre [10,11] can produce large bandwidths suitable for few-cycle pulses, but the energy scalability is limited by the nature of the mixing process as well as the achievable stability. Upshifting ultrashort NIR pulses by white light generation followed by optical parametric amplification (OPA) [12] is approaching the few-cycle regime, but the conversion efficiency is low and here scalability is again an issue, as without stretching the amplified pulse the optical damage threshold is quickly reached. Another technique proposed by Kaindl *et al.* [13] consists of two-stage down-shifting of a Ti:Sapphire laser to access the mid-IR wavelength, which can generate even sub-60 fs pulses, but requires — as all other systems mentioned above — intense (mJ), ultrashort (<100 fs) CPA systems as drivers, and is therefore inherently limited in achievable stability and repetition rate. Many applications of ultrafast mid-IR pulses such as ultrafast spectroscopy [14] or photoionisation studies are highly sensitive to intensity fluctuation, and require extremely stable sources able to operate over large collection times.

Optical parametric chirped pulse amplification (OPCPA) offers a different approach, which can avoid many of the above issues while still meeting the demand for high repetition rate, few-cycle pulse duration and CEP stability. OPCPA can directly produce amplified bandwidths supporting few-cycle mid-IR pulses, and it can preserve the CEP stability of these pulses during amplification. It is an inherently energy scalable process, with joule energy pulses already produced. The conversion efficiency is favourable when compared to the total ns pump-to-Ti:Sapphire output-frequency converted light of a traditional mid-IR source. Unlike the gain-storage media used in traditional CPA amplifiers, no energy is deposited in OPCPA, meaning that the limitation of repetition rate lies only with the available pump lasers. Appropriate pump sources are available for a large range of pulse durations and output energies: Femtosecond systems with significant pump pulse energy typically employ CPA which makes the resulting OPCPA system equally complex as direct gain-storage CPA lasers

and less reliable than long pulse master oscillator power amplifier (MOPA) systems. Nanosecond durations on the other hand require large temporal stretch factors for the OPCPA process and high energy pump lasers. Picosecond MOPA systems present a good compromise in terms of readily achieving pump pulse intensities for OPCPA whilst requiring moderate seed stretch factors and avoiding the complexity of CPA.

State-of-the art picosecond high-energy pump lasers are commercially available in a wide range of configurations with excellent performance characteristics. With the pump laser's stability having direct influence on the stability of the OPCPA its jitter becomes an extremely important parameter, especially in light of achieving better signal to noise measurements with higher repetition rates and faster cumulative acquisition. Such high repetition rate and faster acquisition will only lead to improved signal to noise for integrating measurements if the stability of the source is high. For example in case of the measurement of the change of absorption in a material as in [15] fluctuations over a few percent already proved a limitation during the experiment. As of today, OPCPA sources have achieved stabilities from 1.5% and higher [16,17] while solid state lasers perform on a better level.

2. Experimental setup

Our OPCPA is built with a picosecond high-average power pump laser from Lumera Laser GmbH. The pump laser operates at 1064 nm with 100 kHz at 40 W output power and with pulse duration of 8 ps. Its spatial mode is close to $M^2 \approx 1.2$ and it has stability better than from the most advanced Ti:Sa CPA systems; power fluctuations of <0.4% pulse-to-pulse and <0.1% RMS over 15 hrs are routinely observed. Part of the layout has been described previously [18] and the full system is shown in Fig. 1. The seed for the OPCPA is derived from a two-color fiber laser system (Toptica Photonics) which delivers amplified and phase-coherent ultrashort pulses at 1050 nm (48 fs, 16 mW) and 1550 nm (75 fs, 180 mW). Basing such a system on a fiber seeder has the tremendous advantage for everyday operation that it operates completely hands-off without the need for realignment. The fiber oscillator is used as master oscillator and the

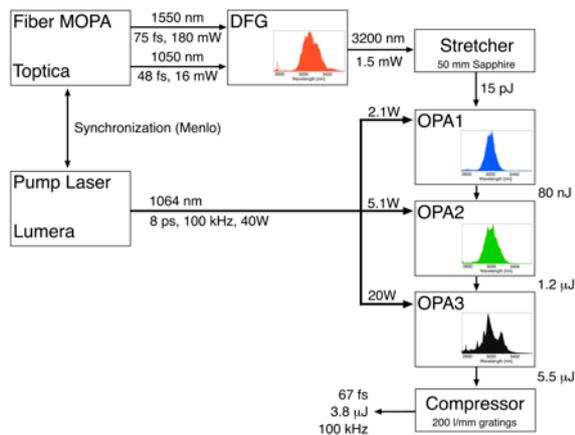


Fig. 1: Mid-IR OPCPA source layout. The two-color output from a commercial fiber MOPA system (FFS, Topptica Photonics) generates, via DFG, self-CEP stable, 3.2 μ m radiation. These pulses are then stretched and amplified by a triple stage OPCPA pumped by a Nd:YVO₄ laser (SuperRapid, Lumera Laser) and finally compressed by a Martinez-type compressor. The compressor includes a linear deformable mirror with which we fine tune dispersion.

pump laser's oscillator is slaved to it to better than 350fs rms over 6 hours via an electronic synchronization unit (Menlo Systems).

Central to the OPCPA is its difference-frequency generation (DFG) stage: DFG between the two frequency shifted pulses from the fiber system allows generation of an ultrabroadband seed pulse in the mid-IR spectral region [19]. Moreover, since the input is derived from two phase-coherent pulses that originate from the same fiber oscillator, DFG yields self-CEP stable pulses without the need for arduous and complicated locking electronics. There is also no need for octave spanning oscillators nor seed bandwidths and as a consequence complexity is reduced significantly [20]. DFG is achieved with a simple, 2 mm long, periodically poled lithium niobate crystal (PPLN), which yields a sub-picosecond duration mid-IR pulse with a spectrum covering 400 nm of bandwidth at the FWHM level with a power of about 1.5 mW at 100 MHz. The PPLN crystal is poled in a fan-out fashion as to allow fine-tuning in the mid-IR; the spatial variation of the fan-out poling is however chosen to vary slowly enough in order to avoid noticeable spatial chirp across the generated mid-IR beam. Previous simulations show that the mid-IR pulse is already negatively chirped [21] and a 5 cm long block of undoped Sapphire is sufficient for further negative stretching.

The stretched mid-IR seed pulse is difficult to characterize temporally since its energy content is only 15 pJ. We estimate the ratio between the pump duration and seed duration to about 4:1 from changing their timing overlap in the first OPCPA stage and monitoring the spectral shift and idler energy. Amplification is achieved in 3 successive OPCPA stages with similar crystals as for the DFG, which we describe in the following: The choice of operating parameters for the first stage is crucial since the highest gain is achieved here and the pump power is distributed in such a way to limit the gain in each stage to prevent super-fluorescence and reversion but also to keep the bandwidth as broad as possible for compression. We refrain from operating at maximum possible gain in order to achieve a good balance between amplification and parametric fluorescence background. The gain in each stage is respectively, 8×10^3 , 40 and 10 with the spatial ratio in the crystals between pump and idler of 3:2. It should be noted that no measurable super-fluorescence was observed in this condition of the layout. Changing the delay between pump and seed pulses to remove the temporal overlap results in no measurable super-fluorescence, and blocking one arm of the fibre laser always brings the power of the mid-infrared wavelength to a value under the minimum measurable which corresponds to a drop of at least 4 orders of magnitude. Operating at the above-mentioned gain values requires a pump power of 2.1 W at 100 kHz for the first stage and results in an amplified idler (note that our seed is the idler wave) bandwidth of 200 nm FWHM, with 80 nJ energy for a pump intensity of 60 GW \cdot cm⁻². Dielectric mirrors ensure that pump and signal waves are rejected and do not participate in any of the following amplification processes. The second OPCPA stage is pumped with 5.1 W at a pump intensity of 57 GW \cdot cm⁻², which results in an idler energy of 1.2 μ J with 250 nm bandwidth FWHM. 20 W of pump are used in the last booster stage which, when optimized for maximum energy conversion results in 1 W of average power or 10 μ J per pulse in the idler centered at 3.1 μ m. Taking advantages of the fan-out design of the crystal we can optimize for a maximum bandwidth of 350 nm FWHM, the power drops to 550 mW or 5.5 μ J. The maximum bandwidth implies a reduction of power as each

stage amplifies a slightly different part of the seed spectrum.

Compression of the negatively chirped pulses is achieved with a simple setup resembling the Martinez-type stretcher typically employed in the visible to near-IR range. We opt for a grating based compression setup since it provides easy power scaling and avoids the high intrinsic losses, and possible thermal problems, of a mid-IR prism compressor setup. Suitable prism materials, which exhibit the necessary positive dispersion, are e.g. semiconductor materials such as Germanium or Silicon with large bulk linear absorption (on the order 3200 cm^{-1}) even if Fresnel losses are removed with proper coatings. We use two gold-coated 200 line per millimeter gratings and place a 1D deformable mirror with a silver-coated membrane (OKO Technology) in the Fourier plane of the 4-f stretcher setup.

The deformable mirror is used to fine tune dispersion as required to generate few-cycle pulses. The compressor setup as a whole is designed to support 600 nm spectral bandwidth and exhibits a measured transmission efficiency of 70% for 350 nm bandwidth FWHM. Second order phase is solely adjusted through its grating separation with the deformable mirror operating passively without any deformation; minimum achievable pulse durations were measured to 85 fs.

It operates with 19 actuators and provides a maximum displacement of $9 \mu\text{m}$, which corresponds to a delay of about 60 fs at our wavelength range. Each actuators range is discretized in 4096 steps or about 2 nm and addressed individually. The mirror is computer controlled and its optimum configuration was obtained by the use of a genetic algorithm to converge to the shortest pulse. As a control measurement we compared the conversion efficiency of the SHG of the compressed pulse to that of the uncompressed one. The algorithm considered a population of 60 individuals, crossover and mutation were included and we took care that no excessively steep gradients were applied to neighbouring actuators across the membrane. In our case the spectrum was spread over approximately 8 actuators only and convergence was achieved after about 180 generations.

3. Results

Figure 2 shows a FROG measurement of the shortest pulses obtained with the above-mentioned procedure and the retrieved temporal profile and spectrum [22].

The shortest measured pulse duration is 67 fs with the transform limit supporting 57 fs. The discrepancy stems from insufficient illumination across all actuators since the compressor was designed for a larger spectral bandwidth. The low frequency fringes at the edges of the frog trace arise from high order spectral phase, and contribute to the side lobe in the temporal profile. The energy at the output of the system for the shortest pulse was $3.8 \mu\text{J}$.

As already mentioned above, such a high repetition rate ultrashort pulse system is extremely attractive not only for strong field physics but for any measurement in which a high signal to noise ratio and cumulative acquisition is important. We monitored the power of the compressed pulse over half an hour by sending the full beam onto a pyroelectric detector from OPHIR (3A head, acquisition rate 3Hz) and measured a power stability of 0.75% RMS over 30 minutes - see also Fig. 3. The stability is excellent and - to our knowledge - signifies the lowest jitter observed from any OPCPA system. We note that our measurement represents a low-pass filtered result due to the inadequate response times of the detector, especially for the 100 KHz repetition rate. Such an analysis is analogue to most measurements quoted in the literature but higher peak-to-peak fluctuations might exist which we plan to investigate in a

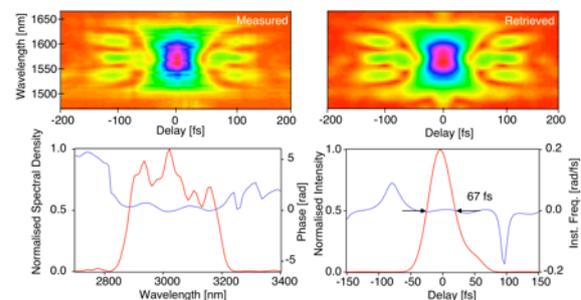


Fig. 2: Measured and retrieved SHG FROG traces of the compressed pulse (FROG Error = 0.41%). Left: The temporal intensity and instantaneous frequency of the compressed pulse. Right: the spectra after each nonlinear stage.

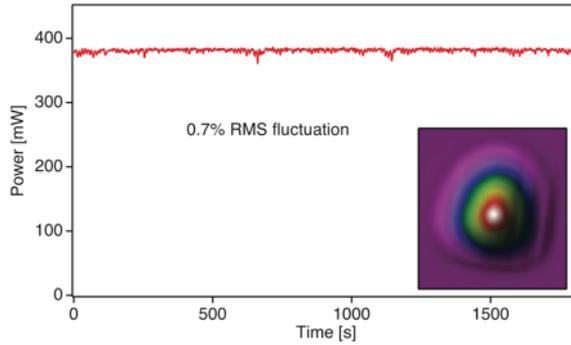


Fig. 3: Stability of the system and profile. The stability of the OPCPA system after compression is excellent with fluctuation under 0.75% over 30 minutes. Insert is the beam profile after compression

future measurement. The insert in Fig. 3 shows a knife edge measurement of the compressed pulse and the high spatial quality, which is sufficient for focussing close to the diffraction limit.

4. Conclusion

In conclusion, we present a source of extremely short pulses in the mid-IR, with high power stability [23]. The compressed pulse energy is $3.8 \mu\text{J}$ at 100 kHz with pulse duration of 67 fs (6.3 cycles) centered at $3.1 \mu\text{m}$, and should already be sufficient for focussing to intensities of $\sim 10^{13} \text{W}/\text{cm}^2$, enough for strong-field physics experiments. The use of a fibre seed oscillator and solid state pump laser contribute to excellent reliability and hands-off operation, while its stability is the highest reported to date for any OPCPA system with 0.75% RMS over 30 minutes.

Acknowledgements

We acknowledge partial support from the Spanish Ministry of Education and Science through its Consolider Program Science (SAUUL - CSD 2007-00013), "Participation in ELI" (CAC-2007-37), as well as through "Plan Nacional" (FIS2008-06368-C02-01). This work is also part of a Collaborative Research Program between ICFO and the Ontario Centres of Excellence, Canada and funding from LASERLAB-EUROPE, grant agreement no. 228334, is gratefully acknowledged.