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Broadband 6-μm OPA Driven by Yb:CaF₂ DPSSL System

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Abstract: 6-μJ, 6-μm pulses compressible to 1.5 optical cycles are generated in multistage OPA driven by 500-Hz Yb:CaF₂-DPSSL. The system is favorable for verification of the predictions on λ-scaling of photoelectron wavepacket dynamics and atmospheric spectroscopy.

OCIS codes (190.7110) Ultrafast nonlinear optics; (190.4410) Nonlinear optics, parametric processes; (140.3070) Infrared and far-infrared lasers.

1. Introduction
The pursuit of tabletop ultrafast coherent X-ray sources based on higher-order harmonic generation (HHG) faces two fundamental challenges: i) the extension of the photon energy into the multi-keV and eventually hard X-ray range (>12 keV) and ii) the increase of the photon flux. In recent years, the theory predictions on the cutoff photon energy scaling as λ³ of the pump laser wavelength have been successfully put to the test. Moreover, highly efficient HHG was realized by exploiting full phase matching conditions in a high-pressure waveguide and the scaling of the phase-matched X-ray cutoff was found, both theoretically and experimentally, to obey a similar, λ⁻¹.7, pump wavelength scaling law [1]. Recently we have demonstrated the generation of bright soft X-ray radiation with energies above 1.6 keV (λHHG<0.8 nm) by employing a 4-μm pump source based on an optical parametric chirped pulse amplifier (OPCPA) in KTA crystals [2]. According to the theoretical predictions [3] phase-matching in the case of longer wavelength multi-millijoule femtosecond pump sources would allow generation of coherent soft X-rays at even shorter wavelength providing a higher spatial and temporal resolution. However, theoretical HHG treatment becomes progressively challenging with the increase of the optical cycle duration and requires taking into consideration additional effects. The potential complications include i) the contribution of the magnetic field component of a linearly polarized driver pulse (i.e. the Lorentz force or vxB) and ii) confinement of HHG to short propagation lengths due to the effect of plasma defocusing in the case of long-wavelength driver pulses which is expected to significantly diminish HHG yield from high density media [4]. Fortunately, the impact of these and other, unforeseen, effects can be studied even at low pulse intensities, which are sufficient to ionize the target gas but are too low for phase-matched HHG. In this contribution we report on a pilot 6-μm source which will be employed in electron wave-packet dynamics measurements using coincidence momentum and velocity mapping imaging techniques.

Our prototype femtosecond OPA is based on ZnGeP₂ (ZGP) crystal that generates broadband 6-μJ sub-200 fs pulses. The measured pulse bandwidth sustains FWHM 30 fs pulses, which is less than two optical cycles at 6 μm wavelength. The required intensities above 0.5×10¹⁴ W/cm² necessary for ionization wavepacket imaging can be reached by modest focusing.

2. Results and discussion
Extending the OPCPA wavelength above 4 μm requires switching to dedicated mid-IR nonlinear crystals and IR short pulse lasers for pumping the OPA because of the IR absorption losses and low quantum efficiency of a near-to mid-IR frequency conversion. Because of good optical quality, broad phase-matching bandwidth and high nonlinearity ZGP is one of the most attractive crystals for the generation of ultrashort pulses in the mid-IR spectral range. Due to the detrimental absorption the ZGP crystal should be pumped at a wavelength above 2 μm [5], that is achievable either by Ho- or Cr-based lasers, or by parametric frequency down conversion of Nd- or Yb-based laser light using an OPA. The latter approach is demonstrated here. Both in direct >2-μm pump laser as well as in indirect >2-μm pump generation approaches, all-optical synchronization of the apparatus is desired which means that the seed for both, pump and OPA, should originate from a single source (Fig.1). The requirements for the frontend are as follows: i) It should generate sufficiently short pulses to facilitate efficient and temporally compressible white light (WL) generation in the visible spectral range and in the spectral range 1.4-1.7 μm. These WL continua are required for seeding OPA1 and 2 which produces, respectively, the seed and pump pulses for the last, ZGP, OPA3 cascade. ii) The frontend should provide high pulse energy for the parametric amplification of the WL to ensure
suitably high energies at the input of OPA3. iii) The frontend wavelength should be long enough to ensure high quantum efficiency of frequency conversion into the mid-IR and match the transparency window for the pump/signal/idler frequency triplet in suitable nonlinear crystals.

The frontend of the system presented here is a 500-Hz repetition rate cw-diode-pumped Yb:CaF$_2$ CPA system producing 6-mJ, 190-fs centered at 1030 nm [6]. The output of the laser system is split for the generation of 2.1-µm pump and 3.2-µm seed pulses. Since with the 1.03-µm source direct WL generation at 2.1 µm is rather challenging, instead, we opted for an OPA pumped by the second harmonic (515 nm) of the Yb:CaF$_2$ source which produces 2.1-µm pulses by amplifying 680-nm WL generated in a 4-mm thick sapphire plate. The generated 2.1-µm seed is amplified in two subsequent OPA stages based on type I BBO crystals by pumping directly with the 1.03-µm pulses. During the amplification of the 2.1-µm “idler” pulses, now used as a seed in the two 1.03-nm pumped BBO cascades, complementary 2.02-µm pulses are generated simultaneously. The combined energy of the 2.0X-µm outputs is 0.4 mJ. 3.2-µm seed pulses are generated by amplifying 1.5-µm WL generated in a 6-mm thick YAG crystal.

In order to produce a broad seed spectrum in the OPA we have used two independently tunable in-line positioned 6-mm thick KTA crystals. Generated 3.2-µm seed pulses were matched in space and time with the 2.1-µm pump pulses in a 12-mm long ZGP crystal (type I, eeo interaction, $\theta = 57.1^\circ$). Because of the partial transmission of the dichroic mirror which was reflecting seed and transmitting pump, only 160 µJ of pump energy was used. The total output of the ZGP OPA was estimated at 17 µJ, which corresponds to approximately to 6 µJ for the idler (6 µm) and 11 µJ for the signal (3.2 µm) pulses.

The spectra of amplified 3.2-µm and generated 6-µm radiation are shown in Fig. 2. The measured bandwidth of the idler wave centered at around 6 µm spans an octave. The spectrum is strongly modulated due to the absorption of air. The spectrum supports 32 fs pulse duration which corresponds to a 1.5 optical cycle pulse at the carrier wavelength.

In conclusion, by employing a femtosecond Yb:CaF$_2$ amplifier and a multistage optical parametric amplifier chain we have generated 6-µJ pulses with an ultrabroad spectrum around 6 µm and compressible below two optical cycles. The demonstrated Yb:CaF$_2$-pumped multistage OPA is also of interest for atmospheric sensing and as a prospective frontend of a multi-millijoule mid-IR OPCPA pumped by a high energy picosecond Ho laser.

3. References