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14. ABSTRACT

A major challenge in developing attosecond X-ray light sources is to increase the pulse energy at high repetition rate and high photon energy. Prof. Zenghu Chang developed a carrier-envelope phase stabilized, high repetition rate (1 kHz), and short pulse (12 fs) laser system centered at 1700 nm at the University of Central Florida. A high energy (100 mJ), high repletion rate (1 kHz) Nd:YLF laser at 527 nm, as well as a set of transmission-type pulse compression gratings were purchased by the DURIP funds. They have been used in the driving lasers that boosted

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Final Report: High Energy Attosecond Source for Studying Electron Correlation

ABSTRACT

A major challenge in developing attosecond X-ray light sources is to increase the pulse energy at high repetition rate and high photon energy. Prof. Zenghu Chang developed a carrier-envelope phase stabilized, high repetition rate (1 kHz), and short pulse (12 fs) laser system centered at 1700 nm at the University of Central Florida. A high energy (100 mJ), high repletion rate (1 kHz) Nd:YLF laser at 527 nm, as well as a set of transmission-type pulse compression gratings were purchased by the DURIP funds. They have been used in the driving lasers that boosted the photon flux of attosecond X-rays in the "water window" to the level that is sufficient for attosecond photoelectron spectroscopy and attosecond transient absorption spectroscopy at the Carbon K-edge. Strong MIR pulses covering 2 to 4 micron have also by generated by using the DURIP equipment, which seed driving lasers for generating keV attosecond X-rays. Zenghu Chang is sharing the advanced attosecond light sources with his collaborators to carry out the research proposed in the Multidisciplinary University Research Initiatives Program: Post-Born-Oppenheimer Dynamics Using Isolated Attosecond Pulses. It provides unique opportunities to study electron correlation with kilohertz attosecond X-rays experiments in the new spectral range.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received		Paper
04/11/2017	2	Gao Chen, Eric Cunningham, Zenghu Chang. Attosecond pulse generation isolated with an asymmetric polarization gating, Journal of Modern Optics, ( ): 1. doi: 1,039,004.00
04/11/2017	3	Yanchun Yin, Jie Li, Xiaoming Ren, Yang Wang, Andrew Chew, Zenghu Chang. High-energy two-cycle pulses at 32 ?m by a broadband-pumped dual-chirped optical parametric amplification, Optics Express, ( ): 24989. doi: 1,039,005.00
04/11/2017	5	YANCHUN YIN, XIAOMING REN, ANDREW CHEW, JIE LI, YANG WANG, FENGJIANG ZHUANG, YI WU,AND ZENGHU CHANG. Generation of 1.8 to 4.2 μm mid-infrared pulses from difference frequency generation in BiB3O6, Optics Letters, ( ): . doi: 1,039,017.00
04/11/2017	4	Yongsing You, Mengxi Wu, Yanchun Yin, Andrew Chew, Xiaoming Ren, Shima Gholam Mirzaeimoghadar, Dana Browne, Michael Chini, Zenghu Chang, Kenneth Schafer, Mette Gaarde, and Shambhu Ghimire. Laser waveform control of extreme ultraviolet high harmonics from solids, Optics Letters, ( ): . doi: 1,039,016.00
04/12/2017	6	Yanchun Yin, Andrew Chew, Xiaoming Ren, Jie Li, Yang Wang, Yi Wu, Zenghu Chang. Towards Terawatt Sub-Cycle Long-Wave Infrared Pulses via Chirped Optical Parametric Amplification and Indirect Pulse Shaping, Scientific Reports, ( ): 45794. doi: 1,039,072.00

TOTAL: 5

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

Received Paper

TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

1. Zenghu Chang (Lectures), "Principles of attosecond technology: generation and metrology," The Frontiers of Attosecond and Ultrafast X-ray Science, Erice, Sicily, Italy, March 19-28, 2017.

2. Zenghu Chang (Invited), "Isolated attosecond pulses in the water window," American Physical Society March Meeting, New Orleans, Louisiana, March 17, 2017.

3. Zenghu Chang (Seminar), "Soft X-ray shines on new attosecond horizon through water window," University of Ottawa, Canada, March 9, 2017.

4. Zenghu Chang (Seminar), "New generation attosecond light sources," Friedrich-Schiller-Universität Jena, Germany, March 2, 2017.

5. Zenghu Chang (colloquium), "Soft X-ray shines on new attosecond horizon through water window," International Focus Workshop with Annual Meeting of the DFG Priority Program QUTIF17, Dresden, Germany, Feb. 26 - March 1, 2017.

6. Zenghu Chang (Invited), "Attosecond X-rays in the Water Window," Southeast Ultrafast Conference, Clemson, South Carolina, Jan. 18-19, 2017.

7. Zenghu Chang, "Attosecond Optics," Short Course, CLEO (Conference on Lasers and Electro-Optics), June 5-10, 2016. San Jose, CA.

8. Zenghu Chang (Seminar), "Advances in Attosecond Optics Frontier," Xi'an Institute of Optics and Precision Mechanics, Chinese Academy of Sciences, May 24, 2016. Xi'an, China.

9. Zenghu Chang (Seminar), "Advances in Attosecond Optics Frontier," Xi'an Jiaotong University, May 23, 2016. Xi'an, China.

10. Jie Li, "Mid-IR OPCPA Laser for Generating Isolated Attosecond Pulse in the Water Window," CREOL Industrial Affiliates Day 2016, March 10-11, 2016. Orlando, Florida.

11. Zenghu Chang (Invited), "Next generation attosecond light sources," The 19th Annual Southeast Ultrafast Conference, North Carolina State University, Jan. 14-15, 2016, Raleigh, North Carolina.

Number of Presentations: 11.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

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(d) Manuscripts

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Number of Manuscripts:

Books

Received Book

TOTAL:

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TOTAL:

Patents Submitted

Patents Awarded

Awards

1. University of Central Florida Trustee Chair Professor, 2016.
2. University of Central Florida Pegasus Professor Award, 2016.

Graduate Students

NAME	PERCENT_SUPPORTED
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Total Number:	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: ......

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:......

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:......

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):......

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Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME

Total Number:

Names of other research staff

NAME	PERCENT_SUPPORTED

FTE Equivalent:
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Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

See attachment.

Technology Transfer

Scientific Progress and Accomplishments

Zenghu Chang

1. Statement of the problem studied

Few-cycle, mJ-level, and carrier-envelope phase (CEP) stable lasers in the short-wavelength infrared to mid-wavelength infrared are of significant interest to the ultrafast community. One of such interests is to produce isolated soft/hard X-ray attosecond pulses with shorter pulse duration and sufficient photon flux. So far, isolated extreme ultraviolet (XUV) attosecond pulses as short as 67 as have been achieved in high harmonic generation (HHG) pumped by spectrally-broadened Ti:Sapphire lasers [1]. Recent development of few-cycle and high-energy driving lasers with center wavelengths varying from 1.6 µm to 2.1 µm has already enabled the generation of the attosecond soft X-ray pulses in the water-window region (280 to 530 eV) [2–4]. In order to further extend the HHG cutoff photon energy and thus produce shorter attosecond pulses, the development of high-energy few- cycle pulses further into the mid-infrared (mid-IR) region is in demand because of the quadratic scaling of the ponderomotive energy with driving wavelength [5, 6]. In addition, such mid-IR laser sources are ideal tools for the study of HHG in solids [7], incoherent hard X-ray generation [8], acceleration of electrons in dielectric structures and plasma [9–11], breakdown of dipole approximation [12], filamentation in air [13], rotational and vi- brational dynamics of molecules [14], time-resolved imaging of molecular structures [15], and strong-field science in plasmonic systems [16].

Currently, most few-cycle long-wavelength lasers are centered around 2 µm or below [17–25], which are mainly achieved by using either optical parametric chirped pulse amplification (OPCPA) or dual-chirped optical parametric amplification (DC-OPA) [26, 27] pumped by picosecond lasers. Most of those lasers are seeded via intra-pulse difference frequency generation (DFG) [17–19, 21, 22, 24, 25]. Intra-pulse DFG [28–30] has several advantages: more than one octave super broad phase matching bandwidth, no jitter between the pump and the signal, and passive stable CEP for the idler since the pump and the signal are within a same pulse. In addition, it would be much desirable to obtain µJ-level DFG energy, which would reduce amplification stages, unfavorable nonlinear processes and superfluorescence in either OPCPA or DC-OPA. Rapid progress has been made on the generation of short- pulse mid-IR laser sources above 3 µm [31–35]. However, the spectral bandwidth has not reached near one octave, which limits the pulse duration to multi-cycle. Near-octave spanning few-cycle pulses near or above 3 µm have not been enabled partly due to lack of broadband seed pulses.

There are two objectives in the research enabled by equipment purchased by the DURIP funds. First, generation of > 5 µJ pulse spanning from 1.8 to 4.2 µm by utilizing cascaded second-order nonlinear processes, which are initiated by intra-pulse DFG in a BiB_3O_6 (BIBO) crystal and simultaneously amplified in the same BIBO by optical parametric amplification (OPA). Second, development of a scheme achieve mJ level two-cycle pulses covering 2.4-4.0 µm with a DC-OPA in MgO-doped LiNbO3 (or KNbO3). In addition, explore the feasibility of producing mJ, sub-cycle 4- 12 µm pulses with an OPCPA in ZnGeP2 (ZGP). We aim to seed both systems with the 5 µJ, 1.8 to 4.2 µm pulses.

2. Summary of the most important results

2.1 Generation of 1.8 to 4.2 μm mid-infrared pulses for seeding DC-OPA and OPCPA

The experimental setup is shown in Fig. 1. Nanojoule-level oscillator pulses (730-830 nm) are stretched to 360 ps in an Offner-type stretcher ings before the pulse energy is boosted to 4 mJ by a home- made 14-pass Ti:Sapphire amplifier system, out of which 2.6 mJ is compressed to 2.2 mJ, 30 fs by a transmission grating pair and is then sent to a hollow-core fiber (HCF) filled with 30 psi neon for white light generation (WLG). The transmission gratings are purchased using the ARO DURIP funds.

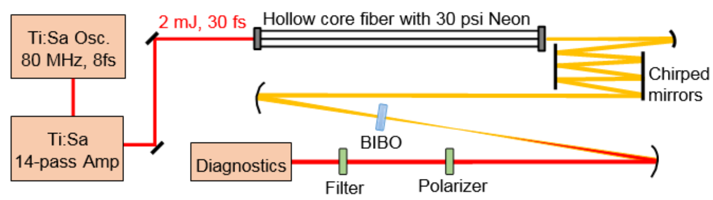

Fig. 1. Experimental setup for the generation of multi-octave spectrum via cascaded second-order nonlinear processes, i.e., DFG and OPA in a single BIBO crystal.

The white light is compressed to ~7 fs using seven pairs of chirped mirrors combined with fused silica compensating plates, after which about 700 μJ energy is usable for experiments. The uncoated BIBO crystal, which has a phase matching angle $\theta = 10.3^{\circ}$ in XZ plane, was placed about 30 cm away from an f=0.5 m concave mirror. The beam after BIBO was collimated with another f=0.5 m concave mirror. The measured two continuum spectra were shown in Fig. 2. In the continuum, the 0.5-0.95 μm spectrum was from the original WLG pulse, which was involved in the DFG process. The 0.95-1.8 μm spectrum was the signal generated from the second nonlinear process–OPA. The 1.8-4.2 μm spectrum in Fig. 2 (a) or the 1.8-3.5 μm spectrum in Fig. 2 (b) was from the DFG process, which was further amplified by the OPA process. The reduced bandwidth in the 0.8-mm BIBO crystal is due to increased group velocity mismatch during the nonlinear processes. A 6.8 μJ pulse covering 1.8-4.2 μm was obtained in a 0.4-mm crystal, compared with a 12.6 μJ pulse covering 1.8-3.5 μm in a 0.8-mm BIBO crystal.

The continuum spectra were generated in cascaded second-order nonlinear processes. The mid-IR pulses are expected to have passive stable carrier envelop phase (CEP), which can be used to seed either a CPA or an OPCPA for achieving high-energy few-cycle mid-infrared pulses

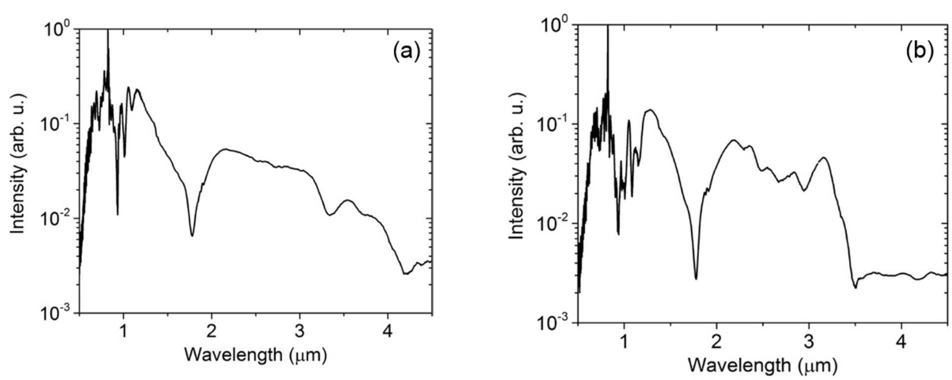

Fig. 2. Measured output spectrum from (a) 0.4 mm and (b) 0.8 mm BIBO crystals with the polarization parallel to the o axis of BIBO crystals, which includes a small portion of HCF spectrum (0.5-0.95 μm), the idler spectrum (1.8-4.2 μm) from DFG, and the new signal spectrum (0.95-1.8 μm) generated in the OPA process when the DFG pulse is amplified.

2.2 Towards Terawatt Sub-Cycle Long-Wave Infrared Pulses

We developed a scheme for not only generating mJ-level, CEP-stable 4–12 μm pulses through optical parametric chirped pulse amplification (OPCPA), but also compressing such pulses to sub-cycle duration through indirect pulse shaping. The μJ-level super broadband spectrum, which covers 1.8–4.2 μm generated by difference frequency generation (DFG) in a BIBO crystal, is used to seed the signal for generating 4–12 μm pulses in an OPCPA pumped by a 2 μm narrowband laser source. The 4–12 μm optical parametric bandwidth is achieved by tailoring the phase matching of ZGP. The second-order phase of high-energy 4–12 μm pulses can be compensated by using NaCl, which has a very small n_2 and a high damage threshold; the higher-order phase can be compensated indirectly by controlling the signal phase with a commercially available acousto-optic programmable dispersive filter (AOPDF).

We simulated the amplification of 4–12 μm idler pulses by an OPCPA in ZGP pumped by a 2 μm picosecond Ho:YLF laser. The approach we proposed is shown in Fig. 3(a). It can not only generate and amplify a broader bandwidth (4–12 μm) but also compress such broadband pulses to the near transform limit. The high-energy 2 μm from a DC-OPA can seed directly a multipass Ho:YLF CPA, avoiding a regenerative amplifier that would complicate the synchronization between the pump and the idler in the 4–12 μm OPCPA.

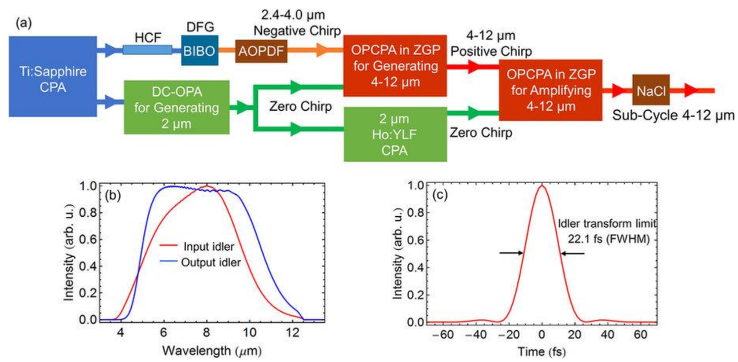

Fig. 3 (**a**) Proof-of-principle schematic setup for amplifying 4–12 μm idler pulses by OPCPA in a 0.7-mm ZGP pumped by a 2 μm Ho:YLF laser; simulation results: (**b**) input (red) and output (blue) idler spectra & (**c**) calculated transform-limited pulse duration (FWHM) of the output idler: 22.1 fs.

The simulation results are shown in Fig. 3(b, c). The input pump centered at 2 μm has a pulse duration (FWHM, transform-limited) of 10 ps and a peak intensity of 20 GW/cm^2. The input idler pulses are stretched to 6.3 ps from 4 μm to 12 μm with a peak intensity of 4.4 MW/cm^2. The input pump-to-idler energy ratio is 10^4. The type I phase matching angle of ZGP is 53°. It can be seen from Fig. 3 that the amplified idler spectrum spans from 4 μm to 12 μm. The conversion efficiency from the pump to the idler is 13.2% (gain: 1317). The compression of 4–12 μm can be achieved by indirect pulse shaping in the idler pulse generation process. The phase of the generated positively chirped 4–12 μm pulse can be coarsely controlled by a NaCl crystal and finely controlled indirectly by controlling the phase of the 2.4–4.0 μm signal pulse using a commercial AOPDF. It is much more favorable to use a short ZGP crystal in the 4–12 μm OPCPA pumped by a 2 μm pump source, which can be achieved by using a short pump pulse at a few picosecond and a high-energy input idler around or above μJ level. Suppose the input idler energy is around 1 μJ, the output idler energy would be 71 mJ with a 600 mJ pump, and the pump-to-idler conversion efficiency would be 11.9%, giving 2.9 TW peak power with a 24.4 fs (FWHM) pulse duration.

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