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14. ABSTRACT
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# RPPR Final Report

## as of 23-Jul-2021

Agency Code: 21XD

Proposal Number: 73891ELRIP

Agreement Number: W911NF-19-1-0224

### INVESTIGATOR(S):

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DUNS Number: 150805653

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**Report Date:** 30-Jun-2021

Date Received: 25-Jun-2021

**Final Report** for Period Beginning 01-Apr-2019 and Ending 31-Mar-2021

**Title:** Single-cycle, carrier-envelope phase locked lasers at 8 micron

**Begin Performance Period:** 01-Apr-2019

**End Performance Period:** 31-Mar-2021

**Report Term:** 0-Other

Submitted By: Ph.D. Zenghu Chang

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**Distribution Statement:** 1-Approved for public release; distribution is unlimited.

**STEM Degrees:** 0

**STEM Participants:** 1

**Major Goals:** The major goals are to develop a high-power long-wavelength infrared attosecond driving laser based on Optical Parametric Chirped Pulse Amplification, which delivers 25 fs (single-cycle), carrier-envelope phased stabilized, 5 mJ pulses centered at 8 micron with a 2 kHz repetition rate. The optical parametric crystals, Zinc Germanium Phosphide (ZnGeP<sub>2</sub> crystals), for amplifying the infrared signal are pumped by a home-build Ho:YLF laser that are driven by Tm: fiber lasers.

**Accomplishments:** Ultrashort intense long-wave-infrared (LWIR) lasers are of great importance to strong-field physics experiments. Scaling of X-ray photon energies by high-order harmonic generation (HHG) to a few keVs requires the development of high-energy few-cycle pulses extending into the LWIR wavelength because the cut-off photon energy of the HHG process scales quadratically with the driving laser wavelength. Furthermore, LWIR lasers are an ideal tool to study HHG in solids, acceleration of electrons in dielectric structures and plasma, breakdown of the dipole approximation and rotational/vibrational spectroscopy.

High-energy femtosecond LWIR pulses can be generated using an Optical Parametric Chirp Pulse Amplifier (OPCPA) or an optical parametric amplifier (OPA) involving frequency conversion in a nonlinear medium. An OPCPA in the LWIR requires the frequency conversion in non-oxide crystal like zinc germanium phosphide (ZGP). However, such crystal is not transparent below 1.9 micron. Hence, the development of high-energy longer wavelength (>1.9 micron) pump sources becomes crucial in the generation of high peak power LWIR pulses. We developed a Ho:YLF Chirped-Pulse Amplification (CPA) laser for pumping a longwave infrared Optical Parametric Chirped Pulse Amplifier (OPCPA) at a 1 kHz repetition rate, which utilizes a Ti:Sapphire laser as a frontend. The 2.05 micron seed pulse is stretched 300 ps by a Chirped Volume Bragg Grating (CVBG). The gain crystals used in the 3-stage amplifiers are 50-mm long, 0.75% doped Ho:YLF cylindrical rods. Each crystal is pumped with a Tm-fiber laser that emits an unpolarized continuous wave at 1940 nm with a power of 120 W. The amplified pulse is compressed using the same CVBG that was initially used for stretching. The measured pulse duration is 4 ps. The CVBG is specially designed by Optigrate Inc. to operate at high energy and is able to handle the peak powers exceeding 3 GW/cm<sup>2</sup> and pulse energies above 50 mJ. With 36 mJ input on to the CVBG, the output energy is 28 mJ. This is the highest pulse energy ever achieved for a kilohertz Ho:YLF laser system utilizing a CVBG for compression.

We developed an OPCPA system that can generate and amplified pulse with a broadband spectrum covering 5.5 to 9 micron. The output of a Ti:Sapphire laser is used to generate mid-IR pulses covering the 2000 nm to 3500 nm region via Intra-pulse Difference Frequency Generation (IDFG) in a 0.8 mm-thick BIBO nonlinear crystal. A bandpass filter is applied to block the spectrum around 800 nm reducing the pulse energy from 0.7 mJ to 0.3 mJ.

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This is done to limit the effects of the filamentation in air introduced when the beam focuses after passing through the BIBO. The generated 2 microjoule seed is positively chirped by an Acoustic Optic Programmable Dispersive Filter (AOPDF) to 3 ps and the energy is reduced to 100 nJ. The three stages OPCPA is pumped by the Ho:YLF CPA system. The output of the Ho:YLF laser is split into three pulses of 0.60 mJ, 5.4 mJ, and 24 mJ energies to pump the first, second and last stage of the OPCPA respectively. A collinear OPA configuration in the first stage to eliminate unwanted spatial chirp of the idler. A long pass filter (LPF) with a 4.5 micron cutoff wavelength is placed after the ZGP crystal to block both the pump and the signal beams. ZnGeP2 (ZGP) crystal is chosen as the nonlinear optical crystal. ZGP is attractive because of its high nonlinear coefficient ( $d_{36}=75$  pm/V), broad transparency extending from 0.72 to 12  $\mu\text{m}$ , and high laser-induced damage threshold (10 GW/cm<sup>2</sup> for 5 ps pulses at 2.05 micron). The spectrum of the OPCPA ranges from 5.5 to 9 micron.

**Training Opportunities:** A postdoc fellow and a graduate student were trained by developing the long wavelength attosecond driving laser.

**Results Dissemination:** The results have been disseminated by publishing papers in peer reviewed journals, book chapters, and by giving presentations at professional conferences, and colloquium at universities.

Zenghu Chang has used the results in teaching Lasers, and Attosecond Photonics courses at UCF, as well as in teaching a short course at CLEO conference.

**Honors and Awards:** Zenghu Chang, UCF Trustee Chair Professor, 2021 (second term).  
Zenghu Chang, UCF College Excellence in Research Award, 2020.  
Zenghu Chang, UCF Research Incentive Award, 2020.

### Protocol Activity Status:

**Technology Transfer:** Patent application for MULTI-MILLIJOULE HOLMIUM LASER SYSTEM. UCF 34270. Filed Aug 13, 2019.

### PARTICIPANTS:

**Participant Type:** PD/PI

**Participant:** Zenghu Chang

**Person Months Worked:** 2.00

Project Contribution:

National Academy Member: N

**Funding Support:**

**Participant Type:** Graduate Student (research assistant)

**Participant:** Fangjie Zhou

**Person Months Worked:** 12.00

Project Contribution:

National Academy Member: N

**Funding Support:**

**Participant Type:** Staff Scientist (doctoral level)

**Participant:** Yi Wu

**Person Months Worked:** 2.00

Project Contribution:

National Academy Member: N

**Funding Support:**

**Participant Type:** Postdoctoral (scholar, fellow or other postdoctoral position)

**Participant:** Krishna Murari

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**Person Months Worked:** 12.00  
Project Contribution:  
National Academy Member: N

**Funding Support:**

**ARTICLES:**

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Volume: 117 Issue: 14

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Date Submitted: 6/25/21 12:00AM

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Publication Location: United States

**Article Title:** Ho:YLF amplifier with Ti:Sapphire frontend for pumping mid-infrared optical parametric amplifier

**Authors:** Krishna Murari, Fangjie Zhou, Yanchun Yin, Yi Wu, Bruce Weaver, Timur Avni, Esben Larsen, Zenghu C

**Keywords:** Ho:YLF, CPA

**Abstract:** We present a Ho:YLF Chirped-Pulse Amplification laser for pumping a longwave infrared Optical Parametric Chirped Pulse Amplifier at a 1?kHz repetition rate. By utilizing a Ti:Sapphire laser as a frontend, 5-?J seed pulses at 2051?nm laser pulse are generated in a Dual-Chirp Optical Parametric Amplifier, which are amplified to 28 mJ pulses with a pulse duration of 5.6 ps. The scheme offers a potential driver for two-color (800? nm and 8??m) high harmonic generation with an increased keV x-ray photon flux.

**Distribution Statement:** 1-Approved for public release; distribution is unlimited.

Acknowledged Federal Support: Y

**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

**Journal:** Nature Communications

Publication Identifier Type: DOI

Publication Identifier: 10.1038/s41467-020-16480-6

Volume: 11 Issue: 1

First Page #:

Date Submitted: 6/25/21 12:00AM

Date Published: 6/1/20 4:00AM

Publication Location: United States

**Article Title:** Attosecond science based on high harmonic generation from gases and solids

**Authors:** Jie Li, Jian Lu, Andrew Chew, Seunghwoi Han, Jialin Li, Yi Wu, He Wang, Shambhu Ghimire, Zenghu C

**Keywords:** Attosecond, water window X-ray.

**Abstract:** Recent progress in high power ultrafast short-wave and mid-wave infrared lasers has enabled gas-phase high harmonic generation (HHG) in the water window and beyond, as well as the demonstration of HHG in condensed matter. In this Perspective, we discuss the recent advancements and future trends in generating and characterizing soft X-ray pulses from gasphase HHG and extreme ultraviolet (XUV) pulses from solid-state HHG. Then, we discuss their current and potential usage in time-resolved study of electron and nuclear dynamics in atomic, molecular and condensed matters.

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**Volume:** 10      **Issue:** 1      **First Page #:**  
**Date Submitted:** 6/25/21 12:00AM      **Date Published:** 5/1/20 4:00AM  
**Publication Location:** United States

**Article Title:** Generation of few-cycle multi-millijoule 2.5  $\mu$ m pulses from a single-stage Cr<sup>2+</sup>:ZnSe amplifier

**Authors:** Yi Wu, Fangjie Zhou, Esben W. Larsen, Fengjiang Zhuang, Yanchun Yin, Zenghu Chang

**Keywords:** Cr:ZnSe, Femtosecond CPA

**Abstract:** Recent progress in high power ultrafast short-wave and mid-wave infrared lasers has enabled gas-phase high harmonic generation (HHG) in the water window and beyond, as well as the demonstration of HHG in condensed matter. In this Perspective, we discuss the recent advancements and future trends in generating and characterizing soft X-ray pulses from gasphase HHG and extreme ultraviolet (XUV) pulses from solid-state HHG. Then, we discuss their current and potential usage in time-resolved study of electron and nuclear dynamics in atomic, molecular and condensed matters.

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**Acknowledged Federal Support:** Y

**Partners**

I certify that the information in the report is complete and accurate:

Signature: Zenghu Chang

Signature Date: 6/25/21 6:12PM

Ultrashort intense long-wave-infrared (LWIR) lasers are of great importance to strong-field physics experiments. Scaling of X-ray photon energies by high-order harmonic generation (HHG) to a few keVs requires the development of high-energy few-cycle pulses extending into the LWIR wavelength because the cut-off photon energy of the HHG process scales quadratically with the driving laser wavelength. Furthermore, LWIR lasers are an ideal tool to study HHG in solids, acceleration of electrons in dielectric structures and plasma, breakdown of the dipole approximation and rotational/vibrational spectroscopy.

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