Development of New Mid-Infrared Ultrafast Laser Sources for Compact Coherent X-ray Sources

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Introduction

In this project, we proposed to develop *laser based* mid-infrared lasers as a potentially robust and reliable source of ultrafast pulses in the mid-infrared region of the spectrum, and to apply this light source to generating bright, coherent, femtosecond-to-attosecond x-ray beams. The use of “conventional” laser materials, rather than more recently developed techniques such as OPA or OPCPA, has important advantages even when the laser media cannot be directly diode-pumped. First and foremost, laser media store energy in an excited state. If the excited state lifetime of the medium is long, the exact shape in time of the pump pulse is decoupled from the gain of the amplifier. Furthermore, gain transitions have little or no dependence on the relative directions of the pump and the seed laser beams or pulses. A pump pulse can be incident on the gain medium from the front or the side, it can have a very poor focusability and still be useable, and the beam being amplified can pass through the laser medium multiple times from multiple directions to average small fluctuations in pump beam profile that would wreak havoc on an OPA process.

Of course, the success of any laser depends on finding a suitable laser medium. The development of the Ti:sapphire laser medium has been one of the most important factors driving ultrafast laser technology, and ultrafast science as a whole. To implement ultrafast lasers and high-power laser amplifier systems at longer wavelengths in the mid-IR, we need a suitable laser material. In surveying work in mid-IR lasers (primarily developed for LIDAR applications), one class of materials stood-out as being the “Ti-sapphire of the mid-IR.” Chromium-doped Zinc Selenide and Zinc Sulphide (Cr:ZnSe and Cr:ZnS) which are closely-related materials that have gain characteristics that are very similar to Ti:sapphire, but simply shifted in wavelength by $\sim 3\times$, with a peak gain at $\sim 2.4\text{ }\mu\text{m}$.

In the phase I project, we investigated chromium (Cr)-doped ZnSe and ZnS crystals as potential gain materials for mid-IR high power ultrashort pulse lasers, at wavelengths $\sim 3\times$ longer than current generation ultrashort Ti:sapphire lasers. We measured temperature dependent absorption and emission spectra to determine the suitability of Cr:ZnSe and Cr:ZnS crystals for cryogenic lasers. Based on the results we obtained in the phase I project, we designed a laser amplifier system to be implemented in the Phase II project.

Figure 1: Absorption and emission spectra of Cr:ZnS at room temperature (red) and 77K (blue) with doping concentration of (a) $3.8 \times 10^{18}/\text{cm}^3$ and (b) $9.4 \times 10^{18}/\text{cm}^3$. RT: room temperature.

Figures 1 and 2 show the absorption and emission spectra of Cr:ZnS and Cr:ZnSe, respectively, at both room and liquid nitrogen temperatures with two different doping concentrations. The crystals (purchased from Photonics Innovations, Birmingham, AL) were grown by chemical vapor deposition and were subjected to proprietary post-growth thermal diffusion of chromium. All crystals are circular disks with about 2mm thickness and without antireflection coatings. They were mounted with a copper mount in a vacuum chamber. ZnSe crystals have a cubic symmetry, which is optically isotropic, while ZnS crystals have a modified cubic structure with a certain degree of hexagonality, which demonstrates birefringence. As pointed out in the literature published by our crystal manufacturer and others, the difference due to birefringence of Cr:ZnS crystals in the absorption and emission spectra does not exceed 10%. Therefore we treated Cr:ZnS crystals as optically isotropic in the first approximation.

Based on our results from the phase I project and the availability of suitable pump lasers today, we chose Cr:ZnS crystals as the gain medium and erbium fiber lasers as the pump laser. The details of the laser system design, which was implemented in the phase II project, are outlined in the Phase II technical objectives.

Technical Objectives

To date, Cr:ZnSe and Cr:ZnS have been studied primarily as a laser material suitable for eye-safe LIDAR applications. There has been some very recent work in employing this material in mode-locked laser oscillators. However, to-date no one has attempted to use this material for the high-power amplification of ultrashort laser pulses. This project aimed to investigate such a laser amplifier, and to integrate such a system with a set-up for generating coherent extreme ultraviolet and soft x-ray high-order harmonic beams. The critical feasibility question was whether it is possible to construct a laser amplifier system with reasonable pulse energy and average power based on Cr:ZnS gain medium. As such, we decided to focus our efforts on the amplifier. Instead of developing a mid-IR mode-locked oscillator ourselves, the seed pulses would be obtained by using an optical parametric amplifier pumped by a Ti:sapphire laser system, which is one of KMLabs' products. We would also purchase commercially available pump lasers instead of developing an ideal pump laser ourselves.

The technical objectives of the phase II project were:

1. To implement a mid-IR Cr:ZnS-based chirped-pulse amplification laser system operating at 200 kHz repetition rate with a pulse energy of several hundred microjoules. The mid-IR laser system would start with a KMLabs standard Wyvern™ ti:sapphire regenerative amplifier system, which has an output pulse energy of 20 μJ at 100 kHz or 5 μJ at 200 kHz with <50fs pulse duration. Pulses from this amplifier would pump an optical parametric amplifier to obtain pulses at ~2.3 μm to seed a two-stage cryogenically cooled Cr:ZnS amplifier, with each stage pumped by a single 100W cw erbium fiber laser. The seed pulses would be stretched prior to amplification by using a standard grating-based stretcher, and recompressed afterwards by using a piece of fused silica, taking advantage of the negative material dispersion at mid-IR wavelengths.
2. Using this laser system, we would demonstrate a coherent ultrafast soft x-ray source with sufficient flux for applications up to photon energies of approximately 400 eV. Important applications in nano-imaging, bio-imaging, and materials and chemical science will be enabled by such a table-top light source. The predicted soft x-ray flux would be high ($\sim 10^8 - 10^{10}$ photons/sec/harmonic) because true phase matching is possible at high pressures using long wavelength driving lasers, simply by adjusting the gas pressure inside a hollow-core waveguide (demonstrated to 2keV with a 4μm drive laser system). The waveguide will also be excellent for guiding the longer wavelength driving pulses over a long interaction length.
3. Supplemental to the Phase II we proposed to attain images of diatoms at 13 and 4nm, demonstrating the capability of the system. Diatoms make an attractive microscopy sample because they are readily available in a variety of shapes and sizes. The dehydrated silica shells make it possible to image internal structure of complex objects, proving the capabilities of our microscope system and phase retrieval algorithm. We expected much clearer images with the much higher flux of the 2.0μm drive laser; in addition, we were to repeat these images at 4nm.

Performance Schedule

First budget year

Month	1	2	3	4	5	6	7	8	9	10	11	12
Purchase the 1 st 100W Er fiber pump laser for the 1 st -stage amplifier	x	x	x	x	x							
Purchase the 1 st cryogenic cooler PT90		x	x	x								
Purchase parts for the OPA	x	x	x									
Purchase mid-IR FROG	x	x	x									
Purchase parts for the stretcher		x	x	x								
Purchase parts for 1 st -stage amplifier		x	x	x	x							
Tune up in-house ti:sapphire regenerative amplifier (Wyvern™)	x	x	x									
Construct the OPA			x	x	x							
Construct the stretcher					x	x						

Design/construct vacuum chamber for cooling the crystal in the 1 st amplifier				x	x	x	x					
Construct 1 st stage multipass amplifier						x	x	x	x	x		
Purchase AO Q-switches								x	x	x		
Construct the pulse cleaner										x	x	
Obtaining data & evaluating the system											x	x

Second budget year

Month	1	2	3	4	5	6	7	8	9	10	11	12
Purchase the 2 nd 100W Er fiber pump laser for the 2 nd -stage amplifier	x	x	x	x	x							
Purchase the 2 nd cryogenic cooler PT90	x	x	x									
Purchase parts for 2 nd -stage amplifier	x	x	x	x								
Construct vacuum chamber for cooling the crystal in the 2 nd amplifier		x	x	x								
Construct 2 nd stage multipass amplifier			x	x	x	x	x					
Purchase parts for the compressor					x	x	x					
Construct the compressor						x	x	x				
Purchase parts for high-harmonic generation (HHG)						x	x	x	x			
Construct/optimize the HHG setup							x	x	x	x	x	
Evaluating and optimizing the system										x	x	x

Milestones

1. The seed pulse source – an optical parametric amplifier pumped by our in-house Ti:sapphire regenerative amplifier, up and running by the end of the 5th month in the first year.

Completed.

2. Finish constructing and aligning the stretcher by the end of the 6th month in the first year.

Completed.

3. Finish constructing the vacuum chamber for cryo-cooling the crystal in the first-stage amplifier by the end of the 7th month in the first year.

Completed.

4. Finish constructing and aligning the first-stage Cr:ZnS multipass amplifier by the end of the 10th month in the first year.

Completed.

5. Finish constructing the pulse cleaner by the end of the 11th month in the first year.

Incomplete, deemed not necessary.

6. Finish evaluation of the first-stage amplifier at the end of the first year.

Incomplete, material problems start to cause long term running issues and damage at higher powers.

7. Finish constructing the vacuum chamber for cryo-cooling the crystal in the second-stage amplifier by the end of the 4th month in the second year.

Completed.

8. Finish constructing and aligning the second-stage Cr:ZnS multipass amplifier by the end of the 7th month in the second year.

Incomplete, again material problems in the first stage at high pump powers prevented progress.

9. Finish constructing the compressor by the end of the 8th month in the second year.

Completed.

10. Finish constructing and optimizing the setup for high-order harmonic generation by the end of the 11th month in the second year.

Incomplete.

11. Finish evaluation and optimization of the complete system at the end of the second year.

Incomplete, amplifier material ultimately failed at high pump powers. In addition a flood at the company destroyed the optical setup which put the program nearly back to the start. All material was repaired or replaced; however, there was not enough time or money left on the program to fully reconstruct the system.

Performance Schedule, supplement

Month	1	2	3	4	5	6	7	8	9	10	11	12
Purchase X-ray CCD camera	x	x										
Qualify 13nm and 4nm XUUS beams	x	x	x	x	x	x						
Design and construct imaging chamber					x	x	x	x	x			
Take images of diatoms									x	x	x	x

Milestones

1. By 5th month, qualify 13nm and 4nm beams for imaging. Check coherence and wavefront distortion, bandwidth, and shot to shot noise.

Incomplete, however construction and optimization continued while Cr:ZnS system problems were worked out.

2. Finish constructing and aligning the imaging chamber by 9th month.

Incomplete, Imaging chamber construction put on hold due to laser failures.

3. By end of year, demonstrated imaging of biological samples (diatoms).

Incomplete, laser problems proved too difficult to complete system.

Phase II work discussion

Cr:ZnS amplifier layout

The material looked quite promising for attaining amplification of ultrafast pulses directly at 2-2.5 μm . In our initial measurements we were able to attain single pass gains of 1.7 to 6, depending on the overlap of the pump and signal. Figure 3 shows our setup of the laser system.

Figure 3. Layout of Cr:ZnS amplifier system.

Our system started with a modified KMLabs Wyvern Ti:sapphire amplifier, which was improved upon under this work and those improvements have been implemented in our products, which is responsible for our largest revenue stream. Those improvements are: Our TEC cooled Ti:sapphire module, improved stretching and compressing, and our “mini Wyvern” small footprint system due for release later this year. The output from the Wyvern are directed to a high repetition rate OPA, developed under this program, in which two commercial sales have resulted. The pulses are converted into a signal (1280nm) and idler beam (2100nm), where the idler is used for the seed to the Cr:ZnS amplifier. The output of the OPA, and tunability are shown in figure 4.

Figure 4. Output power (a) and spectral tuning (b) of 100kHz OPA.

The idler (2100nm) was separated from the signal (1280nm) by a dichroic beam separator and the output was measured with a mid-IR FROG device to measure the spectrum and pulse width. Figure 5 shows the measurement.

Figure 5. Output of OPA idler at 2100nm (a), and measured pulse shape (b)

From figure 5, the bandwidth is 300nm tail to tail, or 140nm FWHM, and the chirped pulse is ~ 150 fs, compressible to 50fs. These pulses were then stretched in a 600g/mm pulse stretcher on a carbon fiber platform, which gave ultimate thermal stability (carbon fiber coefficient of thermal expansion is ~ 0.5 ppm/K, Stainless ~ 10 ppm/K). Stretcher is shown in figure 6.

Figure 6. Carbon fiber stretcher.

Initial measurements started with a single 1550nm, 50W Er:Fiber laser pump from IPG lasers. The pump light is single mode CW, and outputs through an integrated collimator. The beam is then expanded, then focused onto the Cr:ZnS crystal held at 77K, with a beam diameter of ~1mm. Single pass gain measurements yield gains of ~50% at 28.7W of pump light, Figure 7.

Figure 7. Single pass gain measurements with 28W of pump light from Er:Fiber laser.

By doing 3 passes at lower pump powers (<10W), gains of ~6 were attained, however, as the power was increased, thermal distortions dominated the output mode from the amplifier. Figure 8 shows the gain measured just before total crystal fracture.

Figure 8. Gain of 6 in Cr:ZnS, in three pass multi-pass amplifier before damage.

The exact spot on the crystal seemed to be critical in not damaging the coating or the crystal itself, which pointed to a large non-uniformity in the crystal doping, or AR coating. These types of failures plagued the program, and were very pathological in nature. Some crystals were better than others, but eventually all would fail due to exposure. In addition, failure to contact the material to the copper holder of the cryostat, which proved exceedingly difficult, led to thermal distortions which can be seen in the images in figure 9.

Figure 9. Camera image of output beam from amplifier for 29W (a), 35W (b), and 40W (c) of pump power, showing massive thermal distortion.

We constructed the compressor system for the amplifier, however, it was never used due to the amplifier failure. It was based on the same carbon fiber platform as the stretcher.

Hollow core waveguide high harmonic generator (HHG)

The HHG system (KMLabs XUUS) was also constructed, with many improvements. These improvements were related to running at very high pressures (>10 bar) required for phased matched operation at 13nm, and 4nm. Figure 10 shows the XUUS layout. To test the system, we mounted our camera, filter wheels, and beam alignment modules to the XUUS. A 300 μ J, 5kHz, 45fs Wyvern system was used for alignment and generation of high harmonics, to generate 10^{12} Photons/s at the CCD camera. This generation 2 system is available to our current customers, and we have had three commercial sales for these systems.

Figure 10. Layout of new generation 2 XUUS X-ray generator. Red is gas delivery system, blue is the optical system including hollow core fiber, and green is the vacuum system.

Figure 11 shows the actual system that was used in the tests with the Wyvern. The beam profile from the system taken from the camera at video exposure rates can be seen in figure 12.

Figure 11. XUUS HHG setup.

Figure 12. Output mode of XUUS through $0.2\mu\text{m}$ of aluminum.

This system was also designed so that it can run at the higher pressures required for HHG at longer wavelength, to generate higher energy photons near 4nm.

Flood

During the final stages of trying to work out material problems and test new crystals, KMLabs was the unfortunate recipient of a broken pipe causing a flood which decimated the laser system on 15 August 2011. While most of the materials, supplies, and equipment was recovered and repaired through insurance, the lost time and effort was considered too severe to continue the research. Therefore, the system could not be repaired in the time left on the program.

Discussion

While the program was unsuccessful in the main goal of attaining HHG with a high repetition rate ultrafast laser source, there were many successes that steamed from this research program. Following is a list of the program accomplishments:

- 1.) Improved current Wyvern product design, for >100kHz ultrafast Ti:sapphire products, with three commercial sales.
- 2.) Designed new platform Wyvern system for high repetition rates reducing footprint by 50%, and cost by 33%. Product to be released later this year.
- 3.) Enhanced current high repetition rate OPA product, with one commercial sale.
- 4.) Developed carbon fiber stretchers and compressors which have been included in two commercial Wyvern systems.
- 5.) Developed generation 2 XUUS high harmonic generation product, resulting in three commercial sales.
- 6.) Qualified new XUUS with current Wyvern system.

Since this program started, improvements in Chirped Pulse Optical Parametric Amplification (OPCPA) method have addressed this wavelength region of the spectrum nicely, and it is this technology which promises to be the leading technology for ultrafast in the mid-IR due to its versatility in wavelength tunability, ultimate very short pulse durations, and high 100kHz to 1MHz repetition rates. We believe that Cr:ZnS material has many challenges which need to be addressed before very high repetition rate ultrafast sources can be practical based on this material.