Final Technical Report
on
Research and Development of High Energy 2-Micron Lasers
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Abstract

Our research and development of 2-μm femtosecond lasers over the past few years include development of mode-locked Tm:fiber lasers, super-continuum generation starting at 2 μm, and efficiently converting the femtosecond pulses of well-developed 1-μm mode-locked lasers to 2 μm and 4 μm. In this report we briefly overview our past work through this contract on the development of an all-fiber Tm mode-locked laser and super-continuum generation, and early work on half-harmonic generation of 2 μm and 4 μm femtosecond pulses and present our recent work on high-power half-harmonic generation at 2 μm, GaP-based half-harmonic generation at 4 μm, and theoretical studies of simulton formation in half-harmonic generation.

We have achieved more than 65% of conversion efficiency for the half-harmonic generation, and more than 1.7 W of ~30-fs pulses at 2-μm. We have also achieved ~43% conversion efficiency in the second stage of half-harmonic generation using orientation-patterned GaP (OP-GaP) resulting in ~220 mW of ~three-optical-cycle pulses at 4 μm. Since the initial 1-μm laser was a frequency comb source, we could measure the absolute frequencies of the resulting comb at 2 μm and 4 μm and show that cascaded half-harmonic generation of frequency combs was an intrinsically frequency-locked down-conversion process.

Using a split-step Fourier numerical tool, we discovered a new operation regime of half-harmonic optical parametric oscillators (OPOs), where self-consistent pulses are formed in the resonators, enabling extremely high slope efficiencies. Experimental observations match the theoretical prediction, and they pave the way to further enhancement of the conversion efficiency and spectral coverage of half-harmonic generation.

1) Past Work

In the early program years, we reported the first mode-locked, thulium-doped fiber figure-eight laser [1]. The mode-locked oscillator produced 1.5-ps pulses with 63 pJ of pulse energy at a 10.4-MHz repetition rate with a 3-nm bandwidth at a center
wavelength of 2034 nm. After amplification, the pulses were compressed to 370 fs with ~50 nJ of pulse energy. The oscillator could also operate in a square pulse regime, yielding stable pulses from ~100 ps to 20 ns long with ~100 nJ per pulse after amplification.

We also reported a super-continuum spanning well over an octave of measurable bandwidth from about 1.0 to 3.7 μm in a 2.1 mm long As₂S₃ fiber taper using the in situ tapering method [2]. A sub-100-fs mode-locked thulium-doped fiber laser system with ~300 pJ of pulse energy was used as the pump source.

More recently, we have been developing the concept of half-harmonic generation of femtosecond frequency combs. Half-harmonic generation –the inverse of second-harmonic generation (SHG)– is the down-conversion of an optical input by one octave. It has been achieved in OPOs operating at degeneracy in c.w. [3] and femtosecond [4] regimes, and has been effective in enhancing the sensitivity of spectroscopy [5]. Operation at degeneracy is particularly promising because of the broad instantaneous bandwidth of the output [6], the potential for high conversion efficiency, and more importantly intrinsic phase and frequency locking of the half-harmonic output to the pump due to the phase-sensitive nature of the parametric gain at degeneracy [7]. We have been actively pursuing the idea of utilizing this technique to take advantage of the power scalability and short-pulse operation of commercially available 1-μm mode-locked lasers and translate them to 2 μm and beyond. Last year, we showed more than 60% conversion efficiency of half-harmonic generation at 2 μm [8], and cascaded half-harmonic generation of intrinsically phase-locked 2-μm and 4-μm frequency combs [9]. We also showed how half-harmonic generation can be achieved in fiber-feedback [10] and guided-wave [11] configurations.

2) Recent Work

Since September 2015, our work has been focused on three directions: development of a high-power half-harmonic generation system at 2 μm, GaP-based half-harmonic generation at 4 μm, and theoretical understanding half-harmonic generation in the short-pulse regime. Here, we present the summary of the results we have achieved in these directions.

a) High-Power Half-Harmonic Generation at 2 μm

We achieved half-harmonic generation of 19 nJ, ~35 fs pulses from a sync-pumped 2 μm degenerate OPO. The 2.6 W commercial 1-μm laser is converted with an efficiency of 65%. Pulse quality and remnant phase are measured using frequency resolved optical gating (FROG), showing a slight remnant dispersion for the 177 nm pulse with a time-bandwidth-production (TBP) of 0.42.

The setup for the OPO is shown in Fig. 1. The pump laser is a Onefive Origami-10HP, a soliton ytterbium based system. This laser provides an output pulse train of 2.6 W, 70 fs transform limited pulses centered at 1055 nm with a 90 MHz repetition rate (33
The folded OPO cavity is 3.33 m long and uses off-axis parabolic mirrors with focal length 101.6 mm as focusing mirrors. The resulting spot size in the 1 mm MgO:PPLN crystal is ~47 μm. The crystal is poled to 31.8 μm and is at an operating temperature of 60°C. The pump laser goes through a mode-matching telescope before entering the input coupler (M3). After generation in the crystal, the combined 1 μm and 2 μm output of the laser exits through a 65% output coupler (M6). The leakage through the output turning mirrors is sent through a SF10 prism to disperse the pulse before it is measured on a photodiode. The output of this photodiode, combined with the input to the dither mirror (M8) is used to provide cavity feedback through the output coupler and stabilize the cavity length. Since the pulse is dispersed before hitting the photodiode the position of the photodiode can be used as a wavelength selector. This allows a finer tunability of cavity length and the exact position within a peak the OPO locks to, as shown in Figures 2 and 3.

The OPO is first operated in a dither mode where the cavity length is scanned back and forth several microns. The peak structure of the OPO due to the phase locking condition is easily seen in Fig. 2(a), with each peak being spaced by the pump wavelength (1.055 μm = π radians). The peaks highlighted in red are those that are degenerate. Only the degenerate peaks result in a continuous spectrum and a single coherent pulse. They also result in higher conversion efficiencies. Fig. 2(b) shows how the maximum of each peak changes as the input pump power varies. The peak with the largest negative detuning demonstrates a very high threshold, but also a very high slope efficiency. The other degenerate peaks also demonstrate a higher slope efficiency than the non-degenerate peaks. The highest efficiency demonstrated in this OPO is 65.3%, converting 2.6 W of power on the crystal to 1.7 W of 2 μm output (18.9 nJ @ 90 MHz).
Fig. 2- Output power of the OPO in the dither mode. (a) shows the individual peaks as the cavity length is changed. (b) shows the maximum of each of these peaks for different input pump powers. The peaks highlighted in red are the degenerate, short pulse areas of interest, which are exhibiting high slope efficiencies.

To further investigate the structure of the peaks the output of the OPO is put through a monochromator. Figure 3 shows the result of the same scan as Fig. 2(a) but now with the additional information of the wavelength structure of each peak. The degenerate vs non-degenerate nature of each peak is now clearly seen. The peak with the lowest threshold (see Figure 2b) shows clear signs of being overdriven with a resulting unstable spectrum. The two degenerate peaks are relatively clean and easily locked too. The advantage of having the ability to select the wavelength region to lock to is now clearly seen; different regions within a single peak have the highest power at different wavelengths. As the circuit locks to the highest power point on the photodiode this results in locking to a different position within a single peak. The OPO is then locked to the peak with the highest average power and broad bandwidth, the first peak in Fig. 3.

Fig. 3- OPO spectrum as a function of cavity detuning.

The measured and retrieved FROG traces are shown in Fig. 4(a) and 4(b); FROG error was 0.2%. The retrieved pulse width was 35 fs with spectral bandwidth of 177 nm, giving the pulse a peak power of 475 kW. This corresponds to a time-bandwidth product of 0.42. The residual spectral phase was found to correspond 400 fs$^3$ of third-order dispersion (TOD) and no residual group velocity dispersion (GVD), implying that the pulse is close to the transform limit and Gaussian. In order to verify that the FROG measurement is accurate we compare the resulting spectrum with an optical
spectrum analyzer measurement, and calculate the frequency marginal as well as an independent autocorrelation.

**Fig. 4** Measured (a) and retrieved (b) SHG FROG trace from the 2-μm OPO short peak. Temporal (c) and Spectral (d) reconstructions, showing a 35-fs pulse with 177 nm of bandwidth, corresponding to a time-bandwidth product of 0.42
The OSA spectrum shows slightly larger bandwidth than the FROG trace with 182 nm FWHM. It also shows significantly more spectral content on the short side of the spectrum in the 1600-1900 nm range. This content is heavily distorted by the water absorption band and any pulse content there is destroyed and does not show up on the FROG trace. The crystal used in the SHG FROG is a 250 μm BBO crystal. As seen in the marginal calculation, the bandwidth of this crystal is not quite enough to convert the entire pulse with equal efficiency. This explains why the transform limited pulse using the OSA spectrum is 24 fs (TBP of 0.34) while the FROG reconstruction gives 35 fs. An autocorrelation also seems to fit with an ~35 fs pulse, however this autocorrelation is also bandwidth limited. In this case the bandwidth limitation comes from the two-photon absorption of silicon which drops off rapidly around 1050-1100 nm, corresponding to 2100-2200 nm photons from our pulse. The FWHM bandwidth of the pulse extends to almost 2200 nm, causing the autocorrelation to show a longer pulse. We are currently waiting on a thinner BBO crystal to do FROG with an even wider phase-matching bandwidth, which will most likely measure a pulse length of closer to 24 fs at 2 μm.

In conclusion, we have so far, demonstrated a high efficiency (>65%) degenerate OPO with 1.7W of average power and up to 475kW peak power (19nJ). This OPO can be locked to a number of peaks, each with its own spectral content and pulse shape. FROG measurements were shown for one of these peaks, showing a 35fs pulse. Characterization of these pulses can still be improved; with bandwidth of up to 185nm present the transform limited duration should be 24 fs (3-4 cycles at 2μm).

b) GaP-Based Half-Harmonic Generation at 4 μm

Since September 2015, we have also achieved a broadband mid-infrared frequency comb with three-optical-cycle pulse duration centered around 4.2 μm, with up to 220 mW average power, via OP-GaP-based half-harmonic generation pumped by mode-locked fiber lasers.

Broadband mid-infrared frequency combs are becoming increasingly used in applications such as optical clocks, trace gas sensing and stand-off detection, with high
resolution and sensitivities down to ppt-levels [5]. Based on ultrafast fiber lasers and frequency down-conversion using orientation patterned semiconductors, half-harmonic generation of such frequency combs offers an unprecedented level of utility for many applications.

Most recently, a new quasi-phasematched semiconductor - orientation-patterned gallium phosphide (OP-GaP) - has become available from BAE Systems using the all-epitaxial processing technique developed at Stanford [12] combined with optimized hydride vapor phase epitaxy to achieve parallel grating propagation to thicknesses >500 μm. OP-GaP has excellent characteristics for mid-IR parametric frequency conversion including a high nonlinear coefficient ($d_{14}=70.6 \, \text{pm/V}$) and wide transparency range (0.5–12.5 μm). Compared to GaAs, it has a large indirect band gap ($E_g=2.26 \, \text{eV}$), which allows pumping at shorter wavelengths, and also lower group velocity dispersion (GVD) at $\lambda < 5.5 \, \mu\text{m}$, easing the need for dispersion compensation. Low dispersion also results in larger grating periods which propagate more easily to large thicknesses. The first OP-GaP OPOs – pumped with ns 2-μm and 1-μm lasers respectively – have recently been reported [13, 14]. Here we report the first femtosecond OPO based on OP-GaP. Moreover, we demonstrate for the first time highly stable high-energy operation of the frequency combs centered at 4.2 μm via OP-GaP-based half-harmonic generation. The laser system produces pulses of only three optical cycles in duration at up to 220 mW average power at 43% conversion efficiency of 2.1 μm radiation to 4.2 μm. The pulses are intrinsically frequency locked to the output comb of mode-locked Yb:fiber laser.

![Fig. 6- Schematic of the experiment comprising two half-harmonic OPOs.](image)

The cascaded half-harmonic OPOs were synchronously pumped (Fig. 6) by a mode-locked Yb:fiber laser with ∼70-fs pulses at 250 MHz and converted in a PPLN-OPO to 500 mW to 2.09 μm with ∼50 fs pulse width [8]. The second OPO is pumped by the signal output of the first OPO. The concave gold mirrors (M6 and M7) have an ROC of 25 mm. The nonlinear gain is provided by a 0.5-mm long plane parallel OP-GaP crystal with a poling period of 92.7 μm for type-0 phase matching at room temperature, which has anti-reflection coatings for the signal and pump wavelength. A 1-mm thick uncoated ZnS substrate is placed in the cavity at Brewster angle for dispersion compensation, providing round-trip group delay dispersion (GDD) of −20 fs$^2$ and TOD of 2200 fs$^3$ (mirrors neglected). The output coupling of the mid-IR signal pulses is provided by a coated pellicle. The 4-μm OPO is put in a box purged with nitrogen to reduce the effects of atmospheric absorption on the OPO operation. Its position with respect to the 2-μm OPO is chosen to result in close-to-optimal spatial overlap of the resonator modes without requiring extra mode matching optics. The
threshold of the 4-μm OPO is 200-300 mW with 25% output coupling and 26 mW in a high-Q cavity.

The output spectrum of the 4-μm OPO is shown in Fig. 7(a). It is centered at 4.18 μm with the 3-dB bandwidth of ~630 nm, and the -30 dB width of ~2 μm. The double-peaked cut in the spectrum around 4.2 μm corresponds to the CO₂ absorption in the 1.5 m in the beam path from the OPO to the spectrometer. The autocorrelation of the pulses is measured by a two-photon detector and is shown in Fig. 7(b). The pronounced autocorrelation wings are due to the dispersion associated with the CO₂ absorption in the beam path. The pulsewidth is estimated to be ~48 fs, assuming sech pulses, while the simulated pulse width for the measured spectrum assuming no CO₂ in the beam path is ~45 fs. The slope efficiency was measured to be 58% and absolute efficiency reached 43% (Fig. 7(c)). We expect that even shorter pulses can be achieved by optimizing the third-order intracavity group delay dispersion and the dielectric coatings of the input couplers and the AR coating on the crystal.

![Fig. 7- Optical spectrum (a), autocorrelation trace (b), and output power (c) of the 4-μm frequency comb.](image_url)

We achieved what is to our knowledge the shortest pulses (<50 fs corresponding to three optical cycles) centered around 4.2 μm from the highly stable broadband and efficient frequency comb produced via a two-stage cascaded half-harmonic generation in OP-GaP. The combs are intrinsically phase- and frequency-locked to the mode-locked laser pump and are characterized by a high conversion efficiency of over 40% from 2.1 to 4.2 μm, and the highest output power of 220 mW, with nearly 1 nJ pulse energy.

c) Simulton Formation in Femtosecond Half-Harmonic OPOs

We have also studied experimentally and theoretically the dynamical behavior of a synchronously pumped OPO near degeneracy. In steady state we find the properties of the degenerate peaks to be consistent with the formation of simulton-like pulses.

To date, the conditions under which degenerate operation in femtosecond OPOs is obtained are not well understood. We did numerical and heuristic models of the half-harmonic OPO used in [8] and find that degeneracy is enabled by pulse acceleration akin to the formation of simultons discussed in [15, 16].
The half-harmonic OPO consists of a 1mm long Brewster-cut MgO:PPLN crystal phasematched for degenerate optical parametric amplification (OPA) in a ring resonator pumped by a 1045-nm mode-locked Yb-fiber laser (Menlo Systems Orange A) producing 70-fs pulses at 250 MHz with an average power of up to 950 mW. The group-delay dispersion (GDD) of the crystal is partially cancelled by the input coupler (IC) and the air in the cavity. The OPO spectra for several cavity detunings (detuning spectra) are shown in Fig. 8(a). Oscillation is achieved around cavity lengths for which the signal resonates, corresponding to phase shifts of 0 or \( \pi \) on each round trip. \( \Delta L > 0 \) peaks are seen to oscillate with broadband degenerate spectra, while \( \Delta L < 0 \) peaks split into a distinct signal and idler.

The half-harmonic OPO was simulated numerically using split-step fourier methods for the OPA developed in [17] including the full dispersion of the PPLN, but neglecting \( \chi^{(3)} \) effects which have been shown to be small for the cases considered here. We model the cavity using a linear transfer function from the OPA output to input including length detuning, and mirror transfer functions. The simulated detuning spectra shown in Fig. 8(b), are in good agreement with observations.

![Fig. 8](a) Measured and (b) Simulated detuning spectrum in dB relative to the highest peak.

The simulated output signal and pump pulse evolution is shown in Fig. 9(a). for \( \Delta L = 1 \ \mu m \). When the pump is undepleted, the signal experiences unsaturated parametric gain and a group delay of 3.5 fs relative to the pump on each round trip. After 25 round trips, the pump depletes and the signal accelerates to the front of the pump, reaching a steady state with constant energy and timing with respect to the pump. Steady state occurs when the saturated parametric gain equals the round trip cavity loss and the signal repetition rate matches that of the pump. For \( \Delta L > 0 \) the signal pulse must travel faster through saturated OPA than unsaturated OPA.

Propagation in a saturated amplifier has been discussed by Akhmanov [15] and in the context of 'simultons', coupled bright(\( \omega \))-dark(\( 2\omega \)) solitons, by Trillo in [16]. Generalizing to the case of incomplete pump depletion, we find the shift in inverse
group velocity is proportional to the unsaturated gain coefficient, the pulse duration, and the signal pulse energy. Solving for steady state, we find that the signal pulse width should scale with the inverse of the net unsaturated gain. Figure 9(b) compares this scaling with that observed in simulations, showing good agreement in the dependence on pump power. We note that this mechanism cannot enable degenerate operation for $\Delta L < 0$.

We have shown that a nonlinear increase in the effective group velocity, due purely to $\chi^{(2)}$ and group velocity mismatch is adequate to describe the essential pulse shaping mechanisms in femtosecond degenerate OPOs. Extensions to the larger parameter space and further description of 'simulton' effects found in the heuristic model will be pursued and is expected to enable operation of these OPOs with efficiencies more than 70%.

3) Future Goals

We are currently working to develop funding support for the next step of this program with an ultimate goal of a 1-μm wide comb at 8 μm, using a GaAs-based system.

a) Combining OPO and OPA to Achieve Micro-Joule Pulses at 2 μm

We plan to generate multi-μJ sub-100-fs pulses at 2 μm by extending our recent results on generation of multi-nJ sub-50-fs pulses at 2 μm. Our current scheme is based on high-repetition-rate half-harmonic generation followed by low-repetition-rate optical parametric amplification (OPA). The low rep-rate laser operates at 1 MHz with 10-μJ pulses. The high-rep-rate low-energy (~4 nJ) laser output is directed to an efficient stage of half-harmonic generation, which will result in ~2 nJ pulses at 2 μm. This output will be used as the seed for the OPA, which is to be pumped by the 10-μJ output of the 1-μm dual-laser system.
b) Third Stage Cascaded OPO with OP-GaAs at 8 μm

Following our two-stage cascaded half-harmonic generation, we are currently working on building another stage to achieve a 1-μm wide comb at 8 μm, using a GaAs-based system. Given that the dual-comb locking techniques are well-established in the 1-μm wavelength range, the intrinsic frequency locking of half-harmonic generation and its high conversion efficiency open up the opportunity of achieving a mid-IR dual-comb system from a commercially available dual-comb 1-μm system. Such a system will be able to cover a long-sought wavelength range for molecular spectroscopy, i.e. 1.5 to 9 μm.

4) Refereed Journal Publications


5) Presentations

1- E. Sorokin, A. Marandi, P. G. Schunemann, M. Fejer, I. T. Sorokina, R. L. Byer
   “Three-optical-cycle frequency comb centered around 4.2 µm using OP-GaP-based
   half-harmonic generation,” Mid-Infrared Coherence Sources, Long Beach, CA,
   March 2016.

   generation of frequency combs with ~75-fold spectral broadening,” Nonlinear Optics,
   Kauai, USA, July 2015.

3- A. Marandi, K. A. Ingold, R. L. Byer, “Efficient cascaded half-harmonic generation
   of femtosecond frequency combs centered at 2.09 µm and 4.18 µm from a mode-
   locked Yb: Fiber laser,” IEEE Summer Topicals Meeting Series, Bahamas, July
   2015. (invited)

4- A. Marandi, K. Ingold, R. L. Byer, “Quarter-harmonic generation of femtosecond
   pulses at 4.18 µm from a mode-locked Yb: fiber laser,” CLEO: Science and
   Innovations, SW4O. 8, San Jose, USA, June 2015.

   Synchronously Pumped Degenerate Optical Parametric Oscillator,” CLEO: Science
   and Innovations, SW4O. 4, San Jose, USA, June 2015.

6- A. Marandi, “Half-harmonic generation: from broadband molecular spectroscopy to
   non-classical optical computing,” 6th Annual SU2P Symposium, University of St.
   Andrews, Scotland, March 2015. (Invited)

7- A. Marandi, N. C. Leindecker, M. W. Haakestad, T. P. Lamour, K. A. Ingold, K. L.
   Vodopyanov, R. L. Byer, “Sub-harmonic generation of broadband mid-infrared
   frequency combs for molecular spectroscopy,” Progress in Electromegnetics
   Research Symposium, Guangzhou, China, August 2014. (Invited)

8- K. A. Ingold, A. Marandi, C. Rudy, R. L. Byer, “2.09-µm degenerate femtosecond
   OPO with over 60% conversion efficiency and 0.6-W output,” CLEO: Science and
   Innovations, SM2I. 4, San Jose, May 2014.

9- S. Wolf, C. Phillips, A. Marandi, K. Vodopyanov, M. Fejer, and R. Byer,
   “Dynamical Behavior of an Ultrafast OPO near Degeneracy,” CLEO: Science and
   Innovations, CW1B.1, San Jose, May 2013.

10- C.W. Rudy, K.E. Urbanek, M.J.F. Digonnet, R.L. Byer, “Mode-locked Thulium-
    doped All-fiber Figure-eight Laser and Amplifier around 2-µm,” OPIC, Advanced
    Lasers and Photonic Sources, paper ALPS3-4, April 24, 2013.

    supercontinuum generation in tapered chalcogenide fiber from 1.1 to 3.5 µm,”
    Ultrafast Optics IX, paper Tu3.4, March 5, 2013.


References


