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DEVELOPMENT OF A COMPACT BROADBAND OPTICAL PARAMETRIC OSCILLATOR FOR ULTRA-SENSITIVE MOLECULAR DETECTION

by

SEAN O. CRYSTAL

A thesis submitted in partial fulfillment of the requirements
for the Honors in the Major Program in Photonic Science and Engineering
in the College of Optics and Photonics, College of Engineering and Computer Science
and in The Burnett Honors College
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Thesis Chair: Dr. Konstantin L. Vodopyanov

Abstract

Every gas molecule has a unique absorption spectrum that can be captured using optical spectroscopy to identify an unknown sample's composition. Frequency combs systems can provide an extremely broad mid-infrared spectrum that is very useful for molecular detection. A degenerate optical parametric oscillator (OPO) was built to generate the down-converted and shifted frequency comb spectrum. This system utilizes an ultra-short pulse 1.56μm pump laser and a never before used orientation patterned gallium-phosphide crystal. Periodically polled lithium niobate (PPLN), Gallium Arsenide (GaAs) and Gallium Phosphide are all crystals used to accomplish this task. GaP, in comparison to PPLN, has (i) a larger nonlinear coefficient, (ii) much deeper infrared transparency, and (iii) smaller group dispersion – to allow for achieving broad spectral coverage. GaP also has a larger band gap than GaAs; therefore it can still be pumped with a standard telecom C-band laser. An octave-wide spanning frequency comb system was achieved and the characterization of the system is presented. This system is specifically designed to be compact and portable for initial experimental testing in the applications of medical breath analysis and combustion gas investigation.

ACKNOWLEDGMENTS

Table of Contents

Table of Figures

Chapter I: Introduction

Introduction

Every gas molecule has a unique absorption spectrum that can be measured to identify an unknown sample's composition using optical spectroscopy. This process, has existed for many years and is accomplished through various methods. This technique consists of a light source, an unknown sample and a spectrum analyzer. For absorption spectroscopy, which pertains to this thesis, light enters the unknown sample of gas, a portion of that spectrum is absorbed and the remaining spectrum is analyzed to determine the gases composition.

Over recent years, many of the advancements made in this technique have been due to improvements in the light source. This research will focus around the use of a coherent frequency comb light source, with a generated spectrum in the mid infrared, to perform ultra-sensitive gas spectroscopy.

Frequency combs were first introduced in the late 1990s and the technology has evolved significantly since. Now they can span far into the mid-infrared and are produced in many different ways such as mode-locked lasers, difference-frequency generators and, what will be the focus of this study, optical parametric oscillators (OPO). A frequency comb is a series of light pulses from a laser source that consists of many very fine frequency modes with a spacing equal to the repetition rate of the laser [1]. This is a broadband source, which is ideal for spectroscopy. What makes frequency combs unique is that they can produce high power, and most importantly, they are spatially and temporally coherent light sources.

This opens the doors for many applications including national security and forensics, biomedical disease detection, and various industrial applications. The system I am developing is intended for use in biomedical breath analysis and combustion engine efficiency analysis. Exhaled breath containing certain volatile organic compounds such as methane, acetone and isoprene which have been linked to diseases in the endocrine system, lipolysis and cholesterol biosynthesis respectively [2]. Early stage detection of these disease can be very important for successful treatment. It has been shown that using a frequency comb source for spectroscopy allows for ultra-sensitive gas detection that proves useful for early stage detection [3]. Combustion gas engines are the most prominent type of engine and increasing its efficiency significant research is being conducted in this area. The way in which the chemical reaction occurs over time must be analyzed and then can be adjusted to increase the engine's efficiency. Due to the fact that the spectral features of the molecules found in combustion reactions exist over a broad spectrum in the mid-infrared, just as molecules in the breath, frequency combs have recently been proposed to further research in both of these areas to their mutual benefit.

A literature review covering frequency combs generation and ultra-sensitive gas detection will be discussed followed by the intent of our compact OPO system. The equipment that will be used and its purposes are described. The methods of the proposed development of the OPO system and experiments that will be performed are then presented.

Literature Review

Frequency Combs and the Mid-Infrared

A frequency comb is defined as a pulse train of a laser source containing many regular and well defined peaks of laser modes with equal spacing [1]. The comb spacing is equal to the pulse repetition rate (f_{rep}) usually produced by a mode-locked femtosecond laser source. Such spectra

can be used in many application such as ultrafast sciences [4] signal processing and communication [5] and the focus of this paper, gas spectroscopy [1].

Such a broadband source is different than the standard white light source because frequency combs are coherent. The mid-infrared spectral region is ideal for gas spectroscopy because many gas molecules have strong absorption features at these wavelengths, specifically between 3-5µm and 8-13µm where the atmosphere is transparent as can be seen in Figure 1 [1]. We call these absorption features "fingerprints". These strong and defined fingerprints allow us to analyze very sensitive and trace composition of molecules including isotopes.
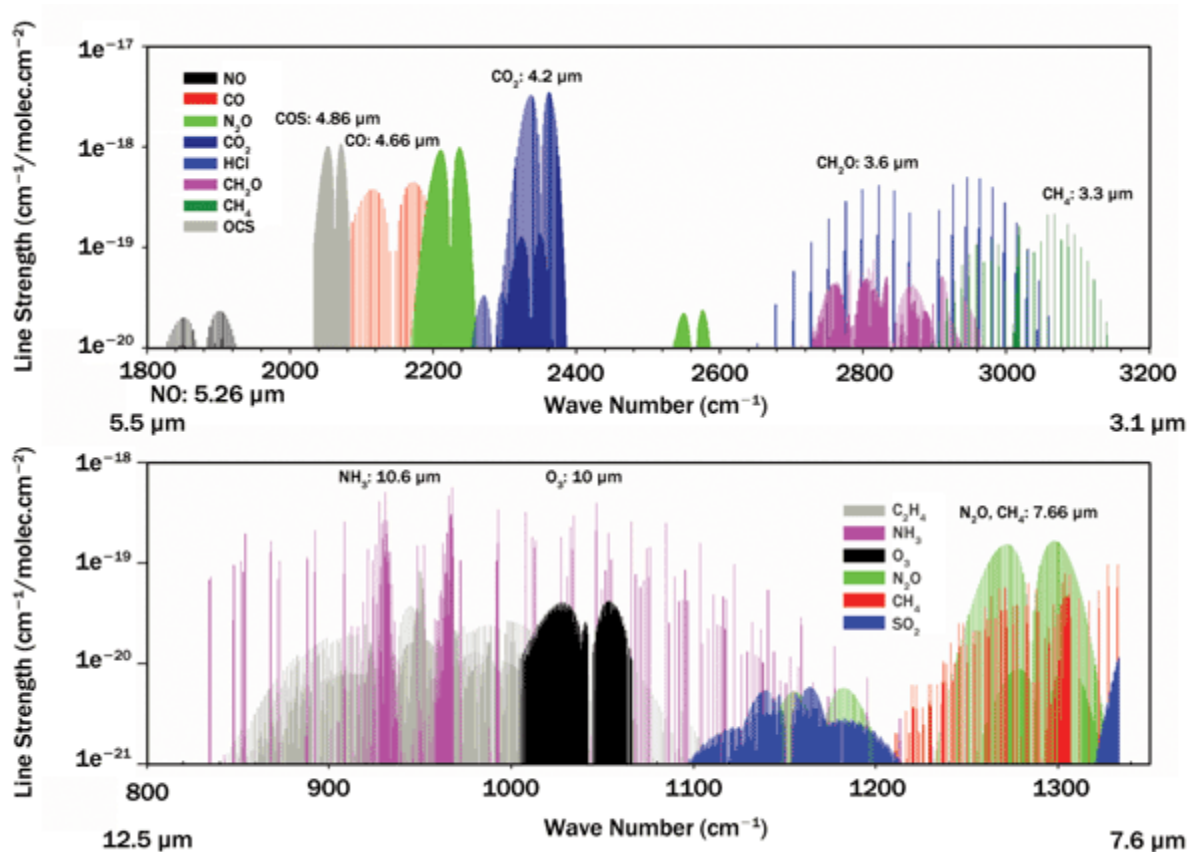

Figure 1: Molecular Fingerprints in the mid-IR [6]

Frequency Comb Generation

The key to all frequency comb applications is the generation of a stable, fine line width and broadband frequency comb spectrum. As mentioned before this usually involves, either as the generator or a pump, a mode-locked femtosecond laser. In mode-locking, a mechanism, either passive or active, causes the various longitudinal modes of the gain medium, and that can also exist in the laser cavity, to have a fixed phase relationship resulting in very short pulses. These very short pulses result in large bandwidth due to the uncertainty principle. When stabilized properly, a femtosecond mode-locked laser can generate a frequency comb [4].

With the current capabilities of mode-locked femtosecond lasers such as fiber doped lasers being limited to near and, in some cases lower mid-infrared wavelengths, there are many methods that utilize the frequency comb mode-locked laser as a pump for other frequency comb generators that go well beyond 3μm. Two popular methods of doing this are through difference frequency generation (DFG) and the use of an optical parametric oscillator (OPO). In DFG, a pump frequency comb of frequency v_{comb} is mixed with a CW frequency v_{CW} and the DFG frequency is then the difference of these two, $v_{DFG} = |v_{comb} - v_{CW}|$ [1]. This process utilizes a nonlinear crystal to produce comb frequencies and the frequency depends on the crystal selected. The efficiency of DFG is dependent on the ability to achieve phase matching. The use of an OPO for frequency comb generation significantly increases the efficiency of conversion in the crystal. Because an OPO frequency comb generator is the method being utilized for the proposed research, the existing literature will now be reviewed in-depth.

As mentioned previously, a femtosecond mode locked laser is the most common choice for pumping an OPO. Lasers ranging from 1μm to beyond 3μm have been used as pumps and the change in pump wavelength and pulse duration effects the output spectrum of the OPO system.

4

Also, depending upon the crystal being used in the OPO, different pump wavelengths will be chosen which will be discussed later. Fiber lasers and amplifiers have become popular for pumping OPOs because of their compactness and good beam quality.

The bowtie configuration, shown in Figure 2, for an OPO is a good option because the oscillating light in the cavity, which is of a different wavelength than the pump, does not pass back through the in coupling mirror and into the pump. The basic bowtie OPO is constructed with a flat dielectric in-coupling mirror (M1) that is usually designed for dispersion compensation followed by two parabolic focusing mirrors (M2 and M3) to focus the light onto the crystal followed by a flat mirror on a piezoelectric stage (M4). For a compact system, such as the one that will be designed, more flat mirrors can be added to fold the cavity.
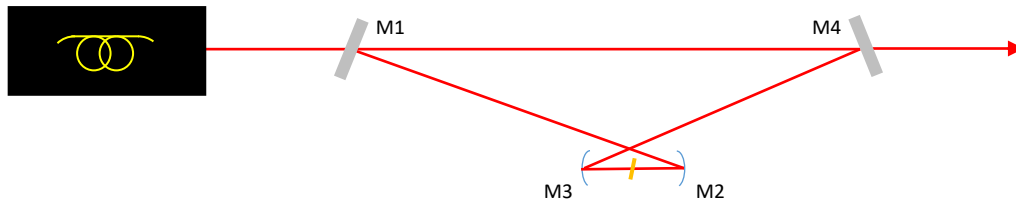
Figure 2: Basic OPO Configuration

The repetition rate of the pump laser determines the size of the OPO cavity. It should be designed so that the OPO length is exactly equal to the length between each pulse in time. This will ensure that there is only one pulse of light in the cavity at a time. It can be deduced that the faster the repetition rate, the smaller the cavity will be. Therefore, for a compact OPO a high rep rate pump laser is very necessary.

The key to converting frequencies in an OPO cavity from the pump frequency to a broad mid-infrared frequency is the crystal between the parabolic mirrors. The beam is focused tightly

down to 10μm diameter onto the crystal. The crystal is periodically poled to be quasi-phase matched for sub-harmonic generation. The crystal is also placed at the Brewster's angle to reduce losses [7]. The pump photons are converted to signal and idler photons with lower frequencies than the pump, respectively, in a divide-by-2 OPO through the sub-harmonic nonlinear process. In the non-degenerate case, the signal and idler frequencies are separate but in the degenerate case they overlap. For most applications the degenerate continuous spectrum case is used but the cavity length can be adjusted to obtain either case. The spacing between frequencies of the idler and signal is equal to f_{rep} [8].

Due to photon energy conservation $\nu_p = \nu_s + \nu_i$, where ν_p is the frequency of the pump, ν_s is the frequency of the signal and ν_i is the frequency of the idler. Their phases are related by $\varphi_p = \varphi_s + \varphi_i + \frac{\pi}{2}$ [8]. The frequency comb is represented by $\nu_m = \frac{f_{CEO}}{2} + m f_{rep}$ and $\nu_m = \frac{f_{CEO}}{2} + \left(m + \frac{1}{2}\right) f_{rep}$ [8]. The value f_{CEO} is the carrier envelope offset and is related to the difference between the phase of the carrier and envelope positions.
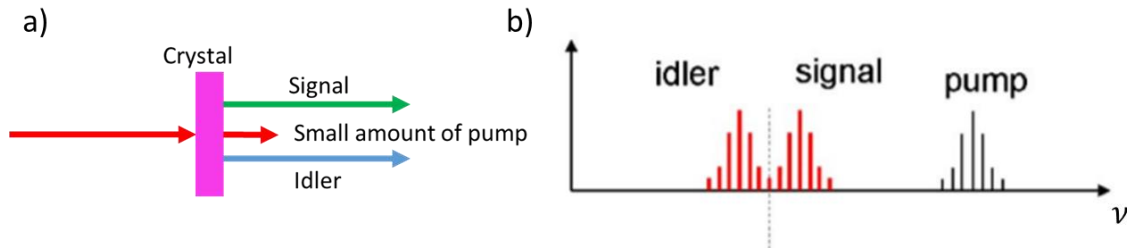

Figure 3: a) Energy representation of photon conversion in OPO. b) Frequency representation of photon conversion in OPO. [8]

Multiple different crystals can be used based on the pump chosen and the, desired output of the OPO. For pumps around 1.5μm periodically polled lithium niobate (PPLN) is popular. For wavelengths greater than 2μm, gallium arsenide (GaAs) has been used.

Spectroscopy with Frequency Combs

Spectroscopy is a science that measures the spectra of elements, molecules or materials based on their electromagnetic properties. There are many different variations of spectroscopy such as emission and absorption. For the purpose of this project we focus on absorption spectroscopy. For example, when light enters a medium of gas, the gas molecules will absorb certain wavelengths of the light based on the particular gas's natural resonance. The light that is not absorbed will pass through and is detected. As mentioned in the introduction, these absorption spectrums are unique to each element, gas, liquid or solid.

This system is comprised of two major components, the light source and the detector. For this thesis we will focus on the light source. There are many different forms of light sources; from incandescent light bulbs, to LEDs to lasers. A major factor in picking a light source is the wavelength that is absorbed by the material being tested. Lasers have become a very popular choice for light sources due to their coherent nature. These lasers typically have very narrow spectra which allows for the high resolution. The downside is that these narrow spectra only allow one to detect a small range of molecules if the center wavelength cannot be tuned over a wide range and even if it can be the range is limited. To see many molecules over a large range multiple lasers would be needed. What to do if you want to detect many molecules over a large range all at once? In addition, what if you need extremely high resolution? This is where the frequency comb comes in as an ideal source for spectroscopy.

Frequency combs produced by OPO cavities can span up to an octave, more than enough spectrum to view many molecules with one source, and provides resolution down to $0.0035\ cm^{-1}$ due to the extremely small line width of the comb teeth [9].

For trace gas detection, the gas to be examined is placed into a cell that is illuminated with the frequency comb source. For best results a multi-pass cell is usually used. A portion of the comb is absorbed and this signal must be analyzed at the output of the system. The Fourier transform technique is used to do this. The output light is sent into a Michelson interferometer, and then a Fourier transform of the interference signal produced is taken to restore the spectrum. The result is a frequency comb spectrum with absorption lines at the resonance(s) of the gas being tested [10]. It has also been shown that intra-cavity frequency comb spectroscopy can be done. In this case the gas is simply placed into the OPO cavity and the output spectrum is then analyzed [11].

Applications

The OPO developed produces spectrums far into the mid-infrared which is making this source very attractive for many applications. Medical breath analysis has begun utilizing this source, and when the system is used as a remote sensor, bomb detection has been explored.

Over 1000 different compounds have been identified to exist in human breath and many molecules have been correlated to specific diseases [12]. While breath analysis is not currently used to officially diagnose such conditions, it provides an easy way to suggest that one should take further tests to diagnose. The reason laser breath analysis is exciting is because it is simple, accurate and completely non-invasive. Most of these biomarkers exist over a broad spectrum in the mid-infrared which is why frequency comb sources are useful. Gases such as CO_2, CO, CH_4 and NH_3, which have links to medical diseases, have more than 8000 spectral features in the infrared [12]. Molecules such as COS, C_2H_4 and CH_2O, which also have links to medical diseases, have strong spectra in the mid-infrared and these compounds are related to very serious conditions

such as respiratory diseases, acute lung transplant rejection, UV-radiation damage and breast cancer [13].

To perform this type of breath analysis, a breath sample is placed into a multi-pass cell as mentioned in the previous section, the frequency comb spectrum is passed through the cell and collected on the other side. Fourier analysis is performed on the collected light and this resolved spectrum is then compared to the HITRAN data base. Extensive work is also now being done to mathematically separate multiple spectral features from a collected spectrum to accurately determine all of its components.

National security has also played a role in developing frequency comb technology. Many chemical components of bombs, such as ammonium nitrate, have spectral features in the mid-infrared. A material of concern can be targeted by a frequency comb and the back absorption reflection can be analyzed to determine if it contains materials commonly used in bombs.

The mid-IR frequency comb is a source and its applications for detection and sensing are vast. Essentially, anything with spectral features in the mid-infrared can be detected by this source. The above applications are only a few of those currently being explored.

Intent

For many of the most important applications of frequency comb Fourier transform spectroscopy, an efficient, compact and portable source that covers the spectral regions from 2μm to 5μm and beyond is needed. This thesis will explore the development of such a system as it applies to the applications of breath analysis and combustion engine efficiency. To accomplish this, a small fiber laser with sub-100fs pulses was selected and a never before used nonlinear crystal, gallium phosphide, was also chosen because it is expected this crystal can produce an

output spectrum of over one octave. These components will make possible a very compact and

efficient system that I expect will obtain the desired broad output spectrum. This system will make

exploring exciting, novel and real world applications for gas analysis possible.

Chapter II: Construction of System

The preceding text will explain/present the construction of the OPO system starting from characterizing the pump laser, designing the cavity, mode matching the pump and the cavity, Constructing the OPO on the optical table, stabilizing the system and purging the system. Many experiments and tests were run to develop the final system. The next chapter will present the results obtained from this system.

The final OPO frequency comb system constructed was built based on the knowledge gathered from two different OPOs pumped by two different Erbium fiber lasers, each with two different crystals (PPLN and GaP). One of these systems, being pumped by the FemtoFiber Pro, was constructed prior to this research and will be adjusted for the current studies. Because the FemtoFiber Pro was pumping an existing OPO system at the start of this study, the pump characterization and cavity design that is presented is that of the FemtoFiber Smart.
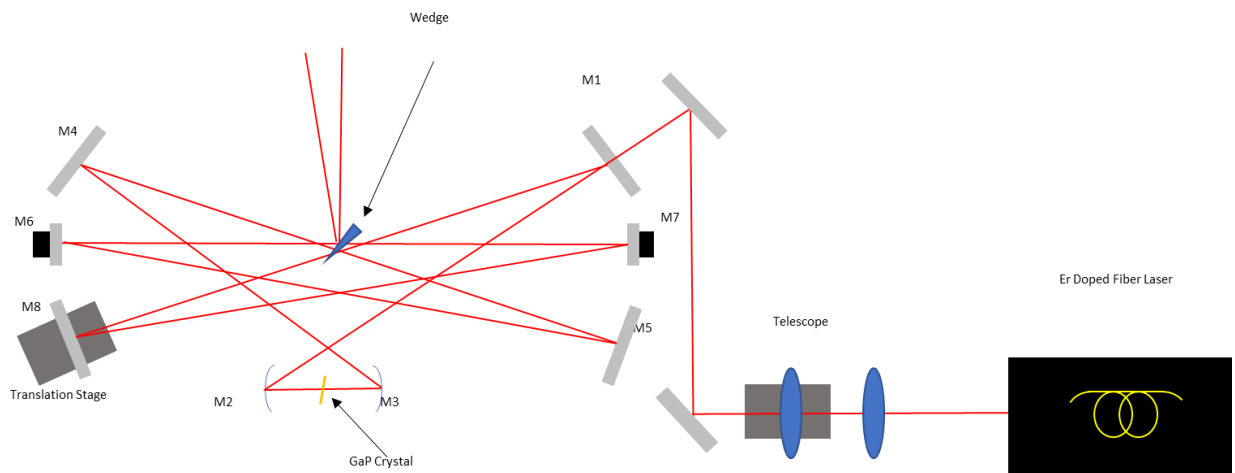
Figure 4: Schematic of GaP OPO system.

Pump Laser Characterization

The characteristics of the pump laser play an important role in how the OPO system functions. Because of the crystals used (PPLN and GaP), a 1.56μm wavelength laser source was chosen. This is a convenient wavelength for a compact prototype system because 1.56μm is a common telecommunication wavelength which means these lasers are highly developed and cheaper than longer wavelength and more exotic pump sources. Both pump lasers, the FemtoFiber Smart and the FemtoFiber Pro, are erbium doped fiber lasers purchased from Toptica Photonics.

The FemtoFiber Smart is more compact and is out coupled through a fiber. The fiber out coupling originally looked to be a unique way to avoid needing a two lens telescope for mode matching, which would decrease the optical path length propagating through dispersive elements, in turn maintaining the shortest pulse duration. This later proved to inhibit the tunability needed to get the proper beam size and divergence therefore a telescope was still needed.

Another main difference between these two pump lasers is the output power. The FemtoFiber Smart has and average power of 140mW and the FemtoFiber Pro has an average power of 300mW. I expected both these lasers average power would be more than enough due to their very short pulse durations (generating a very high peak power). High peak power is needed inside the crystal to produce the desired non-linear effect. It was later found that the FemtoFiber Pro was more useful for this OPO due to its higher average power.

The spectrums of the FemtoFiber Smart is relatively broad. As mentioned above it is centered at 1.56µm. Because this is a mode locked fs pulsed laser, the relatively broad spectrum is to be expected.

Pump Laser Spectrum
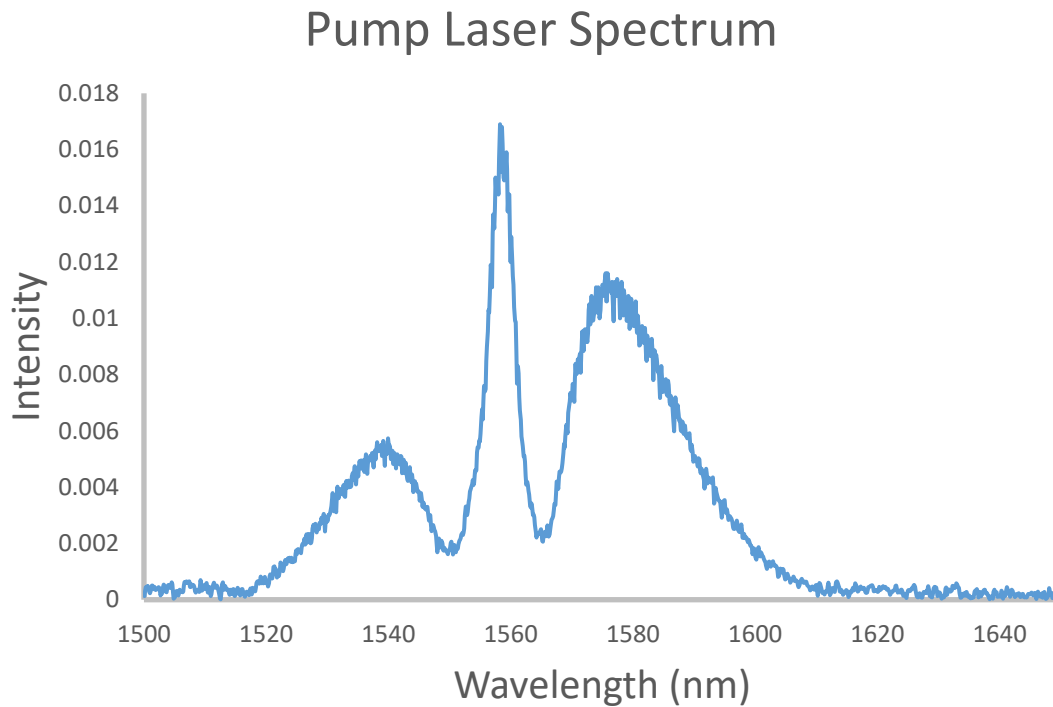
Figure 5: FemtoFiber Smart pump laser spectrum.

To confirm the pulse duration provided by the manufacture, a second order Auto-correlation was performed. This is a widely-used technique if laser science because the pulse duration of lasers play a large role in the performance of many laser systems. This short pulse

duration is a key in this system so this information is very important. A Michelson Interferometric

Second Order Auto-Correlator was built as shown below in Figure 6:
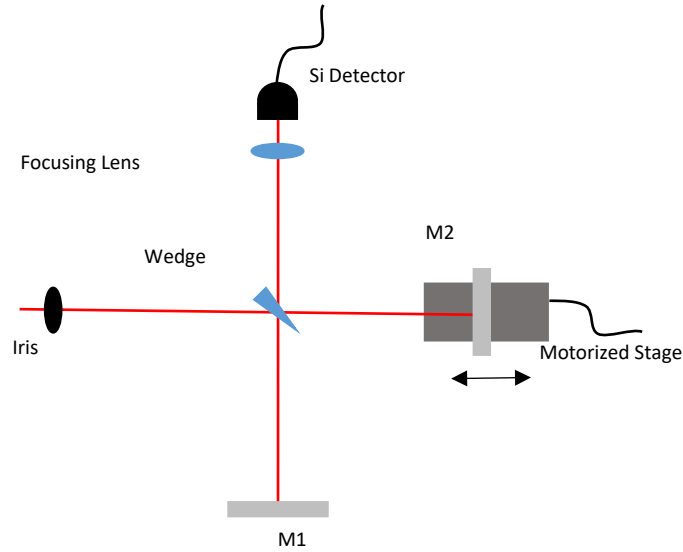
*Figure 6: Second Order Auto-Correlator Schematic
used to measure pump pulse duration.*

The interference signal was gathered and a short program was written in MatLAB to re-

obtain the pulse duration and shape of each interference signal. The results for the FemtoFiber

Smart is shown below in Figure 7 and it confirms the manufactures statistics.
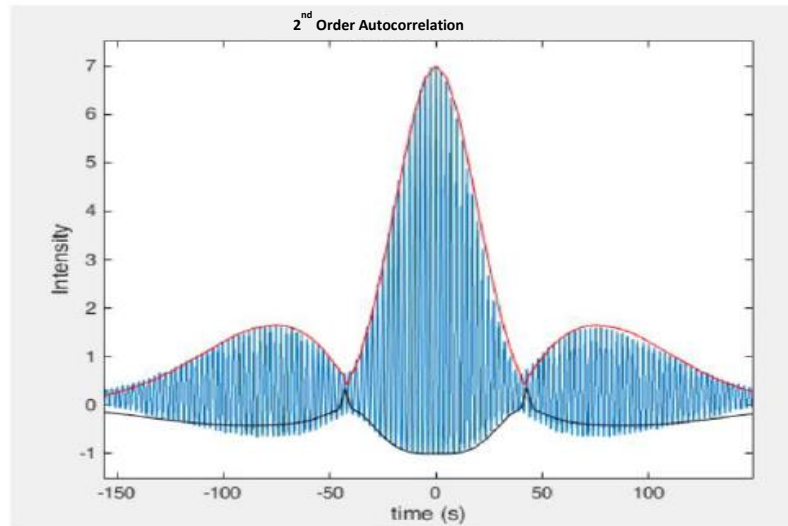
*Figure 7: Collected interference signal (blue) and chirped Gaussian fit (red and
black) of FemtoFiber Smart pump laser.*

14

It was found that the energy of the pump laser pulse is most likeley distributed through a double hump pulse. The first hump of this pulse was found to be between 50fs and 60fs at FWHM. While this partly confirms the data from the manufacturer it also indicates the 50fs pulse duation claimed is true only for the first hump not the entire pulse.

The final data needed from the pump laser is the beam size and divergence so that the OPO cavity can be properly mode-matched. The beam characteristics for the FemtoFiber Smart are provided below in Figure 8 where 0m position will be defined as the in coupling mirror of the OPO for consistency in the later section of mode matching.

Pump Laser Propigation
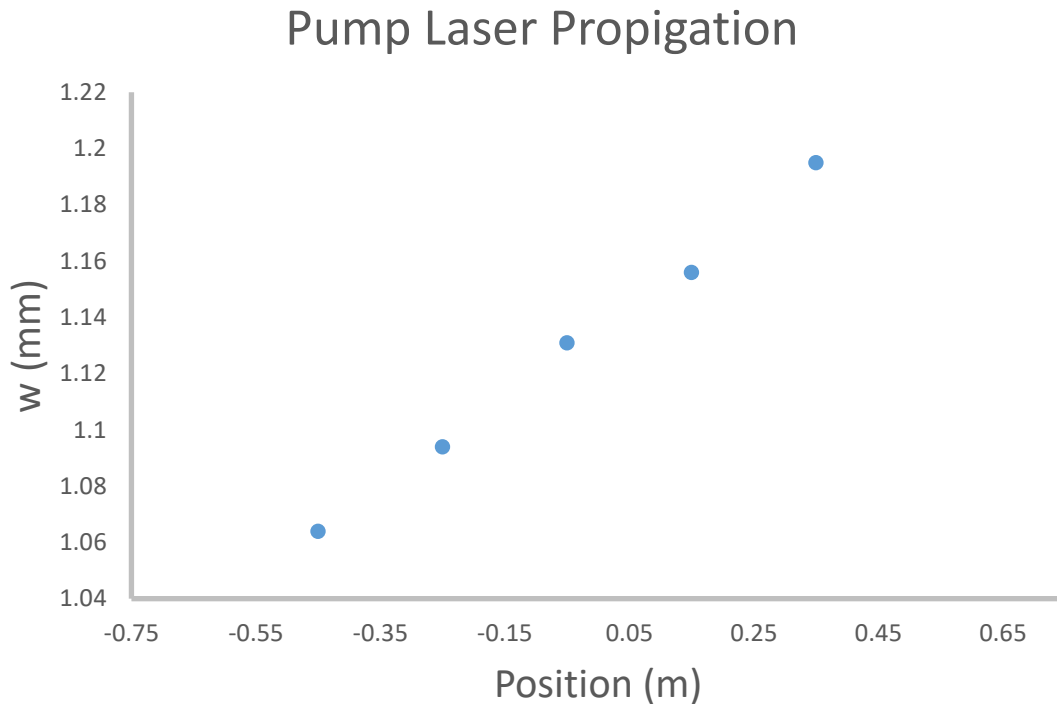
Figure 8: FemtoFiber Smart beam propagation.

<u>Cavity Design</u>

A major design requirement of the cavity is that it must be synchronously pumped and it is ideal that the leak though light of the in-coupling mirror does not propagate back into the pump laser. Due to these requirements, a bowtie ring cavity was chosen (bowtie referring to the beam path resembling that of a classic bowtie). The cavity length is determined by the repetition rate of the pump laser, one pulse enters the cavity at the rate that the preceding pulse leaves the cavity. In the case of the FemtoFiber Pro pump laser, the repetition rate is 80MHz.

$$Cavity\ Length = \frac{3 \times 10^8 m}{s} \times \frac{1}{80MHz} = 3.75m$$

Therefore the overall length of the cavity will be 3.75m. This is a relatively large cavity and to make the system more compact the cavity will be folded.

Figure 9: Eigen-mode of half OPO cavity modeled in BeamSim.

The cavity was simulated in BeamSim. This program allows the Eigen mode of the laser cavity to be modeled. The model, shown in Figure 9, provides information about the stability of the cavity based on multiple parameters such as location of the optical elements and factors of

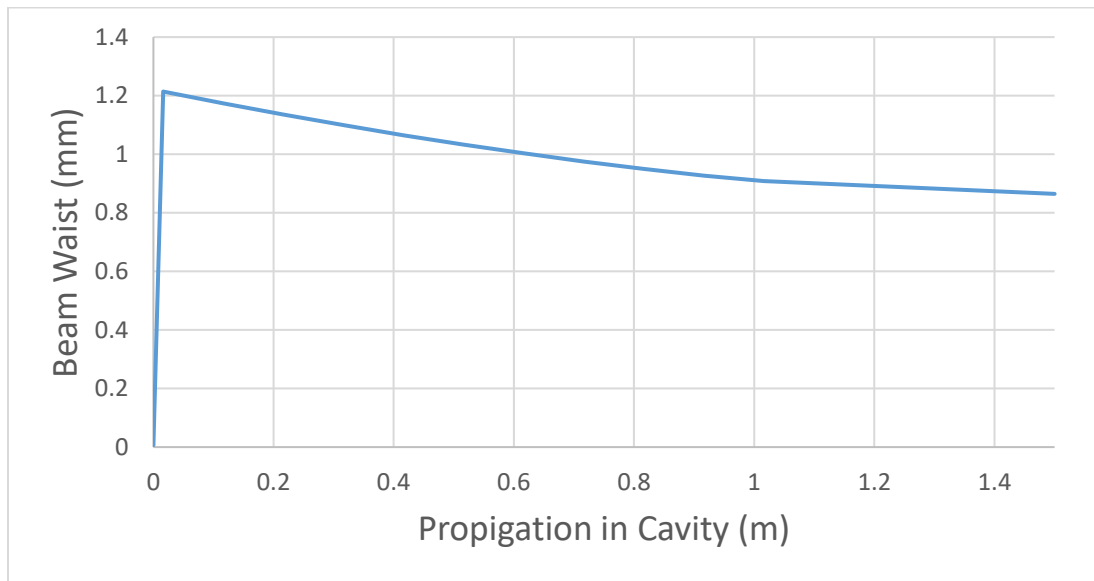the pump beam propagation in the cavity (discussed further in mode matching section). Because the cavity is a ring cavity and ring cavities are symetric, only half the cavity needs to be modeled. In Figure 9, position 1 is the focusing point of the beam inside the crystal, position 2 is at the parabolic mirrors and position 6 is the location of the second beam waist, midway through the cavity.

A schematic of the system was then designed in Solid Works for easy configuration on the optical table. An illustrative schematic of the system can be seen in Figure 4. In order to fold the cavity, 6 flat mirrors will be used instead of the standard two, reducing the size of the cavity to a roughly 1.5ft x 2.5ft rectangle. M1 is an in-coupling mirror that allows the 1.56µm pump to pass though while keeping the broadband 3µm centered idler and signal wavelengths resonating in the cavity. M2 focuses the resonating beam to roughly 10µm inside the non-linear crystal and M3 re-collimates it. A translation stage is used for large cavity length adjustments. Two mirrors in the cavity are attached to one long and one short piezo-electric stage. These piezo's allow for cavity ramping and dither stabilization schemes.

To attain an efficient system, dispersion in the cavity must be controlled. To accomplish this a wedge of dielectric material is placed near the second beam waste. Using a wedge allows adjustment of the resonating beams optical path length through the CaF_2 in order to attain the best dispersion compensation. This wedge doubles not only as a dispersion compensating element but also as an out-coupler. Due to the nature of a wedge two beams will be out-coupled, one will be used for stabilization and the other used for applications.

17

<u>Mode Matching</u>

To achieve efficient and stable lasing, proper mode matching of the pump beam and the Eigen mode of the cavity is necessary. The beam size and radius of curvature of the pump beam must be adjusted to match that of the stable Eigen mode of the cavity described in the cavity design section. A two lens telescope will be implemented where the ratio of focal lengths of the two lenses control the magnified beam size while the distance between them controls the radius of curvature. This second parameter is important because our group has found, in previous systems, that the cavity works best when the beam is slightly diverging. Below, a schematic of a two lens telescope is shown and the equation describing the beam magnification factor is given.

$$Magnification = \frac{f_2}{f_1}\left(1 + \frac{f_1^2}{R_1^2} - \frac{f_2^2}{R_2^2}\right)$$
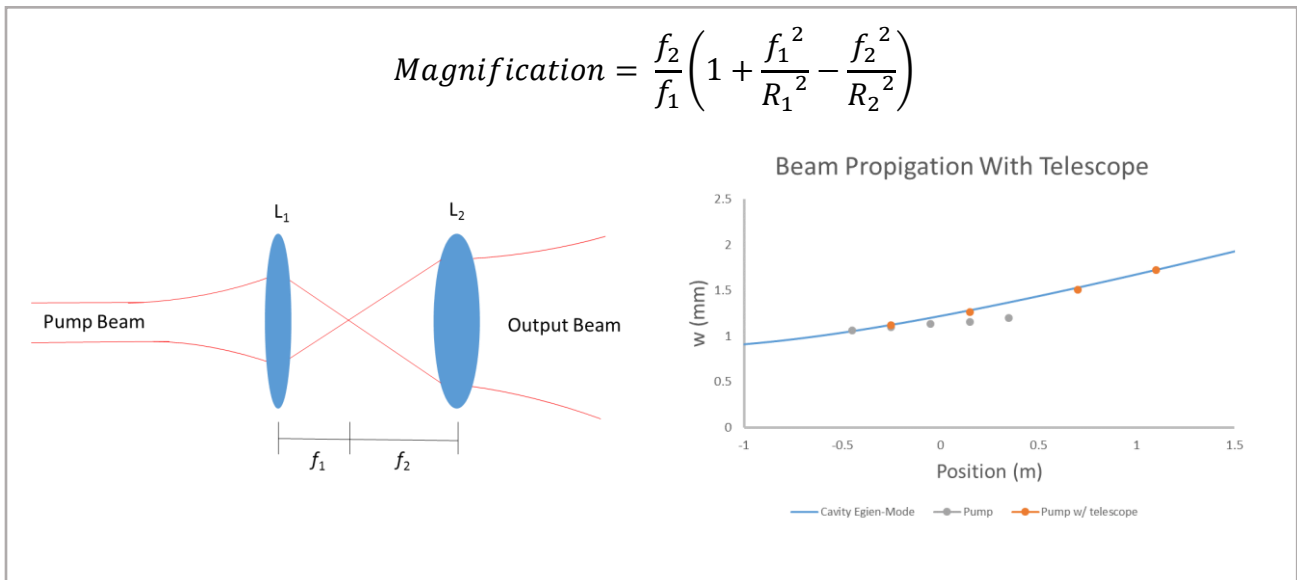
Figure 10: Beam size magnification equation, two lens telescope mode matching technique schematic and Beam propagation plot of cavity Eigen mode, pump beam and pump beam after adding the mode matching telescope.

Data obtained from previous sections, pump beam propagation (Figure 8) and simulated stable cavity Eigen mode (Figure 9) are used to determine the position of the telescope in the propagating pump beam and the needed lenses for proper beam size magnification. Figure 10 shows a plot of the Eigen mode of the cavity with the original pump beam and the pump beam after a proper telescope was implemented, superimposed. The pump beam data in this chart was

collected through a knife edge scan measurement. It can be clearly seen that after the mode matching telescope was added the pump beam propagates along the Eigen mode of the cavity very well.

With the pump beam now adjusted to properly pump the OPO, a cavity can now be constructed that will produce stable lasing.

Construction of OPO

The OPO cavity is constructed based on the information from preceding sections. After the telescope was implemented, an empty mirror mount, which later holds the dielectric in-coupling mirror, was placed at the proper distance from the pump beam. The first parabolic mirror was then placed into the system. Using a thermal camera, the first parabolic mirror position was adjusted until the beam observed in the camera was round and Gaussian. All other mirrors are now placed into position in the order of beam propagation but leaving the in-coupling mirror until last.

Now the cavity length must be adjusted to induce proper synchronous pumping. The accuracy of the cavity length is down to nano-meters. The in-coupling mirror allows the 1.56μm to enter the cavity and anything between 2.5μm and 4 μm will stay resonating in the cavity. But this mirror is not perfectly transmissive or reflective for those respective wavelengths (i.e. a small amount of 1.56μm light will be reflected and a small amount of >3μm light will be transmitted out of the cavity). We take advantage of this imperfection by placing a thermal camera behind the in-coupling mirror and adjusting the cavity length until an interference signal is seen. This interference signal indicates one pulse is entering the cavity as another one leaves (Both impinge on the CCD camera at the same time therefore interfering). The cavity is now synchronously pumped.

At this point the PPLN crystal can be placed between the two parabolic mirrors at the Brewster angle. Its position between the two parabolic mirrors is tuned finely due to the nonlinear processes large dependence on the beam being focused into the crystal. When the position is near optimal a second harmonic visible signal can usually be seen in the cavity.

The cavity is not usually lasing at this point. It will lase at a handful of specific cavity lengths so a piezo is used to dither the cavity back and forth. The translation stage on M6 must now be used to finely adjust the overall cavity length until lasing peaks are detected. When the OPO lases at these specific lasing peaks, the pump beam. This principle is taken advantage of to adjust the cavity to its proper length for lasing. An InGaAs detector replaces the camera behind the in-coupling mirror, and the translation stage is adjusted until pump depletion lines are detected. At this point the InGaAs detector is replaced with a Lead Selenide detector (PbSe) to observe the lasing lines at 3µm. The crystal position and mirror orientations are now adjusted to attain the highest lasing signal.

Finally, an out-coupling/dispersion compensating wedge is added. The wedge allows for a small percentage of light to be extracted from the cavity while keeping enough light in the cavity to maintain lasing. The wedge is rotated to tune the amount of out-coupling. Its position is adjusted to properly tune for dispersion compensation.

After the system is completely characterized with the PPLN, the crystal is swapped for GaP. The same process for placing the PPLN into the system is followed to replace it with the GaP. GaP has different dispersion properties than PPLN so the dispersion compensating wedges must be changed from ZnSe to CaF_2 and adjusted.
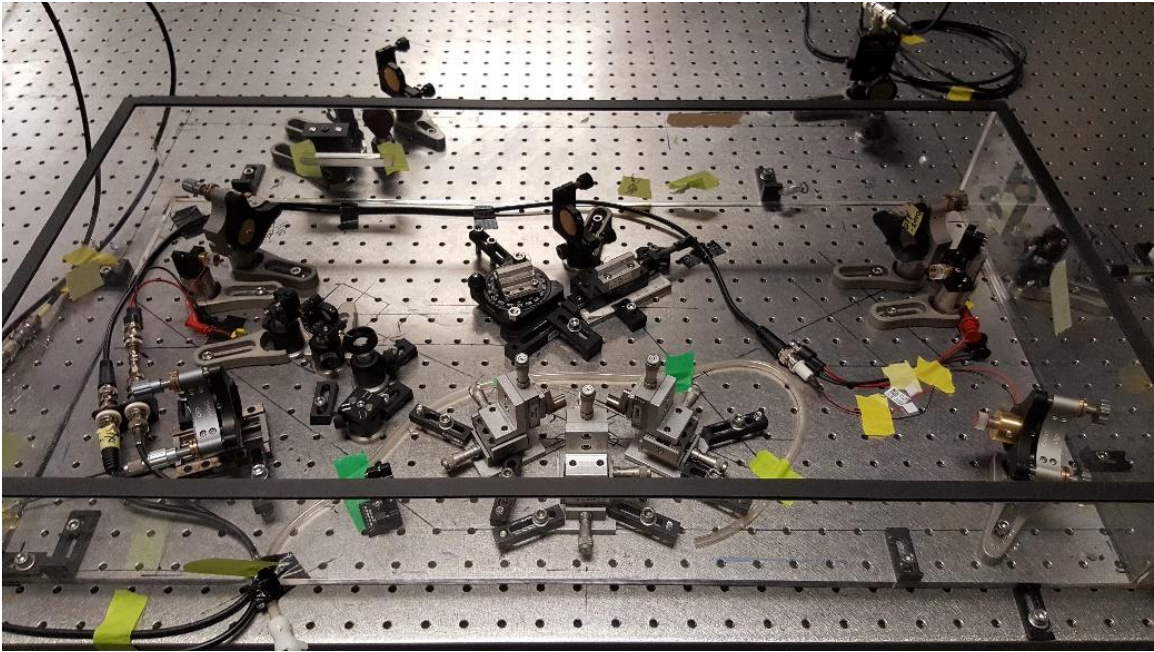
Figure 11: Picture of constructed GaP OPO cavity.

Stabilization

The OPO cavity will lase at select cavity lengths, therefore the cavity can only be operated in two modes, ramp and stabilized. Ramp mode is simply ramping the cavity to shorter and longer lengths (through the length of the cavity that will lase) so that all lasing peaks will lase once through the course of each ramp. While this is useful for construction and many measurements of the cavity, the stabilized mode is more often used for spectroscopic applications.

To stabilize the cavity to a single lasing peak a dither and lock control loop is implemented. For the OPO discussed in this thesis, both an off the shelf lase lock system and individual components (Lock in amplifier, piezo driver, oscilloscope, etc.) were tested. We found that the digital lase lock system performed very well and it was a simpler device to use.

This stabilization technique is based on finding derivative of the lasing peak and sending an error signal to a piezo on a mirror to keep the derivative equal to zero. This can be made more efficient if the lasing peak is made significantly narrow. An approach that was implemented to accomplish this was implementing a grating before the stabilization detector to decrease the bandwidth the detector sees. This worked very well and new non-dither techniques are soon to be explored.

Purging

Water (H_2O) and Carbon Dioxide (CO_2) both have absorption features that lie within the operating wavelength range of this OPO. These features need to be eliminated to attain a good continuous spectrum. In order to accomplish this, a sealed box made of plexiglass is built to house the OPO. Holes are cut for beams and alignment tools. Originally, a constant dry air source was pumped into the box. While this does work very well for water lines, it does not eliminate the CO_2 lines. To accomplish this, the dry air was replaced with Nitrogen gas which eliminates the presence of both H_2O and CO_2.

Chapter III: Results

Spectrum

The most important and unique parameter of this laser system is its extremely broadband mid-IR output spectrum. Once properly dispersion compensated the highly non-linear GaP crystal in the doubly resonant OPO produced a spectrum over an octave wide.
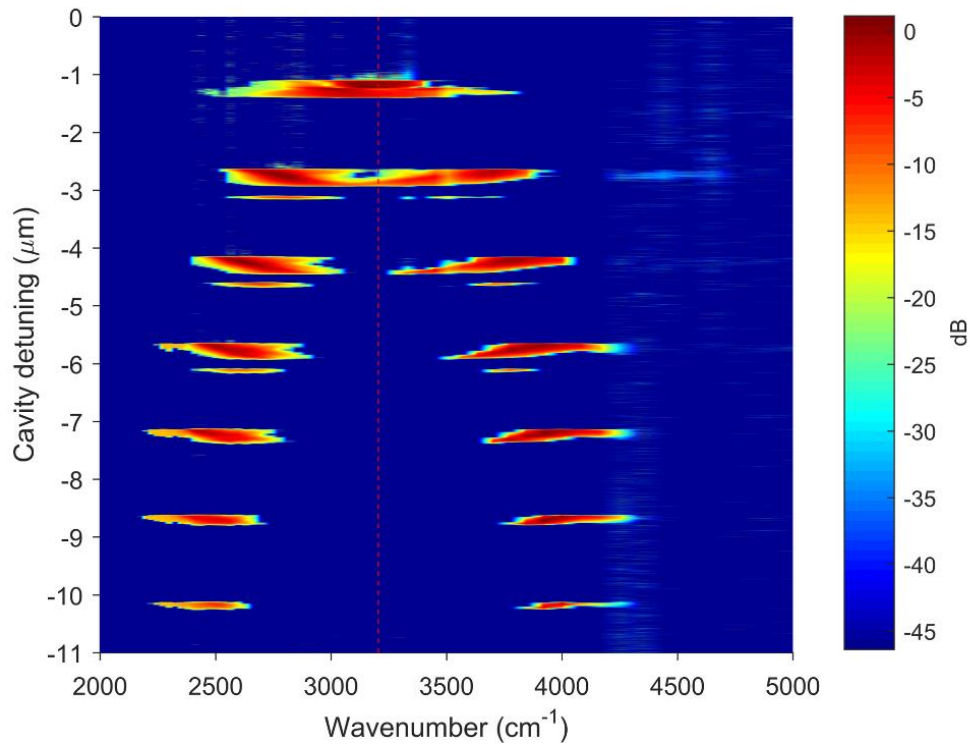

Figure 112: 2D plot of all seven lasing peaks from completely degenerate to completely non-degenerate.
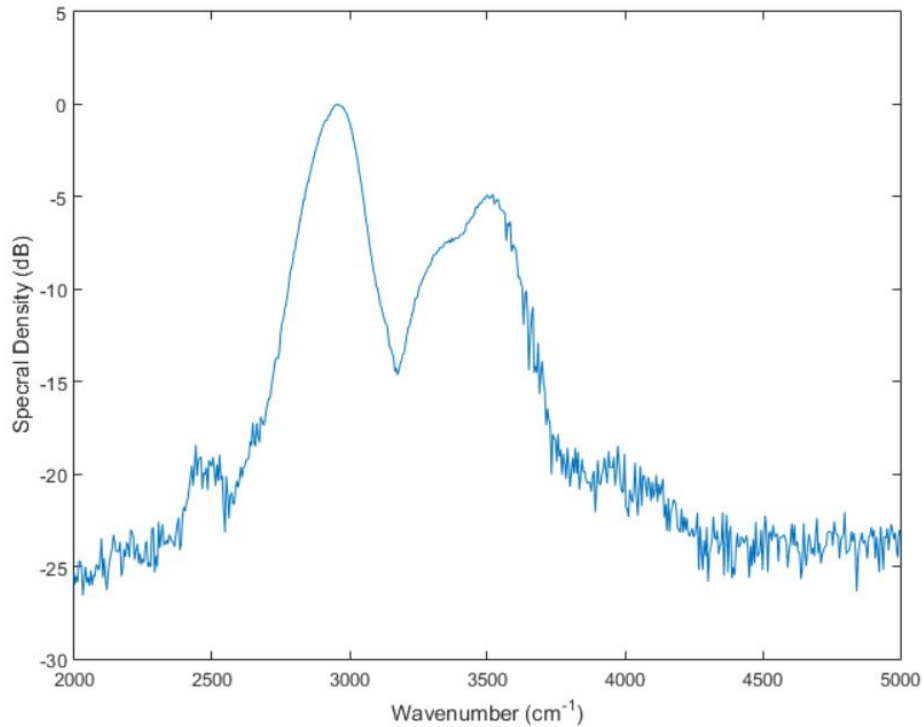
Figure 123: Instantaneous continuous octave wide mid-IR spectrum from OPO. 1D plot of second lasing peak.

Figure 112 presented is a 2-Demensional graphic of the spectral density per wavenumber at each of the seven lasing peaks. This graphic shows how the spectrum varies per peak from completely degenerate (cavity detuning -1µm) to completely non-degenerate (cavity detuning -10µm) as the cavity length is detuned. This data was acquired with a monochromator and PbSe detector. The second peak is the broadest continuous peak but if further mid-IR wavelengths are desired the cavity can be detuned to a more non-degenerate peak.

The second figure is a 1-Demenstional plot of the broadest continuous peak, the second peak. It can be seen that an instantons continuous spectrum spanning 2110 cm^{-1} – 4250 cm^{-1}, greater than one octave, at -24dB, was attained. This is the broadest spectrum attained from an OPO pumped by a standard c-band laser.

The OPO was purged with dry nitrogen for these measurements reducing the concentration of H_2O to 400 ppm and CO_2 to 5 ppm. Even with this, portions of the spectrum were still absorbed around 2300 cm^{-1} and 3750 cm^{-1} due to the minute presence of these molecules.

<center>Outcoupling</center>

As mentioned in the previous chapter, a CaF_2 optical wedge was used to outcouple a small percentage of the laser light from the OPO. Due to the nature of the wedge, two beams are outcoupled. One of these was used to stabilize the OPO while the other will be used for applications.
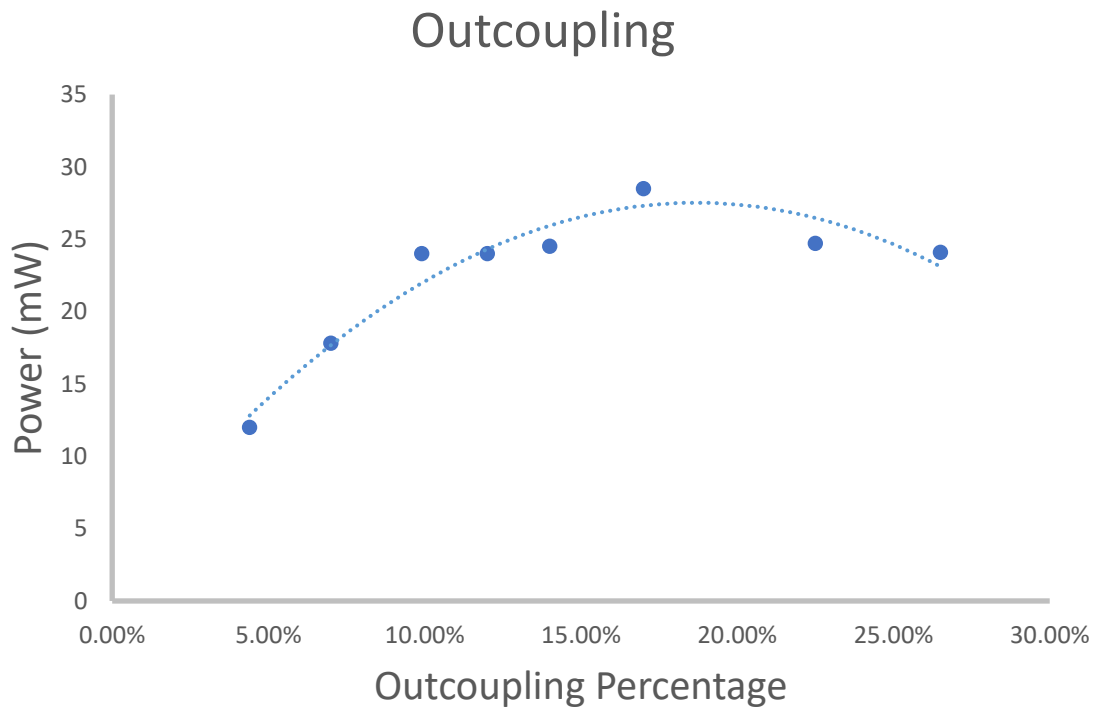

Figure 14: Outcoupling efficiency of OPO at varying outcoupling percentages.

Figure 14 shows the average output power of the OPO vs. the percentage of outcoupling. The outcoupling percentage is increased by rotating the optical wedge closer to normal. This will

also slightly increase the optical cavity length and the translation stage on M6 is adjusted to compensate. The measured output power is the combined power of the two outcoupling beams.

Eight data points were collected and fit with a parabolic curve. It can be seen that the peak output power was measured to be 29mW. This occurred between 15% - 20% outcoupling and at a pump power of 300mW. After this point the output power decreases. Once a certain percentage of light is outcoupled from the cavity, the losses inside the cavity increase significantly and the overall power of light inside the cavity drops off which explains the drop in output power after the peak point.

<div align="center">Cavity Loss</div>

Internal cavity loss is an important parameter in characterizing the efficiency a laser. If the cavity loss is to large the system will be unable to operate much over the threshold, if at all, and will produce very little output power. Since this system will be used for application testing, the efficiency is important.
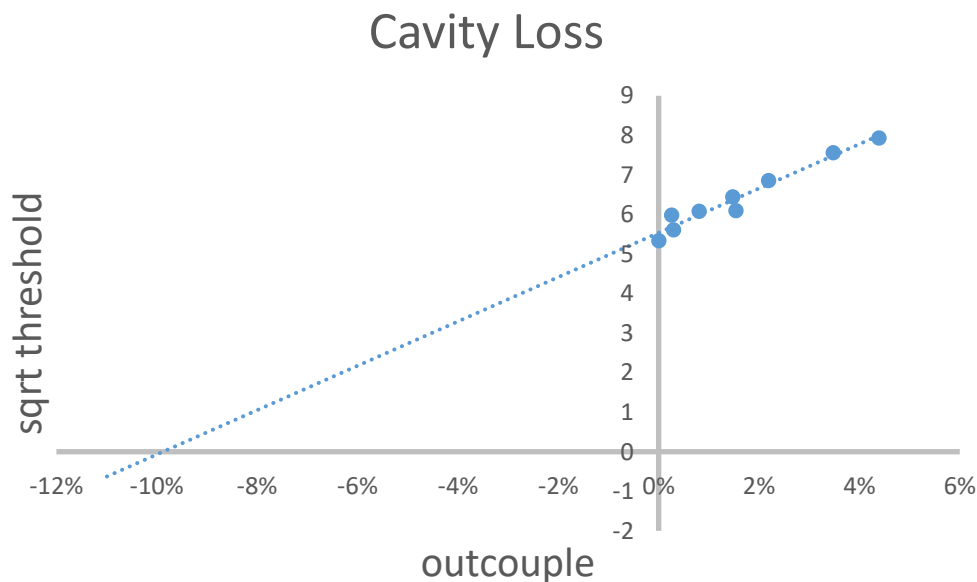

Figure 135: Linear fit of sqrt(threshold) vs. outcoupling percentage, collected experimentally, to determine internal cavity loss.

The cavity loss is experimentally determined by measuring the lasing threshold at varying outcoupling percentages. The lasing threshold is defined as the point where the gain of the laser medium, GaP, reaches the total of all losses in the cavity: $R_{total}e^{(2g_{threshold}l)}e^{(-2\alpha l)} = 1$. The outcoupling percentage is varied by rotating the outcoupling wedge and the lowest pump power that will allow lasing (threshold) is recorded.

$$P_{threshold} = e^{gl}e^{-\alpha l}$$

The lowest threshold value attained from this system was 14mW due to the high non-linearity of GaP. After plotting the sqrt(threshold) vs. outcoupling percentage a line can be fit to these data points. The square root is plotted because the power is the square of the field. The x-intercept of this fitted line is the solution of the loss term in the threshold equation.

Figure 135shows the experimentally collected threshold vs. outcoupling percentage data. As expected, the data point fit closely to a line. After a linear fit was applied and extrapolated to the x-axis, the cavity loss was determined to be roughly 10%. This is a reasonable cavity loss and indicates the system is working efficiently.

<u>Autocorrelation</u>

Interferometric autocorrelation is a technique used in laser science to estimate the pulse duration of a laser. If you know some basic information about the laser pulse, (i.e. if it is Gaussian, Lorentzian, etc.) one can model pulses interfering to match the recorded second harmonic interferogram and determine the pulse duration.

The standard technique used to gather the interferograms is that descried in the Laser Characterization section of Ch.II. While this technique worked well for the pump laser it did not work as well for determining the pulse duration of the OPO output because the OPO output is

significantly lower in power leading to a very low signal to noise ration and the OPO output is very sensitive to dispersion. The solution developed was to generate the second harmonic signal with a non-linear crystal vs. creating the second harmonic signal on the detector itself, like was done for the pump laser. A schematic of this modified second order auto-correlator set up can be seen below:
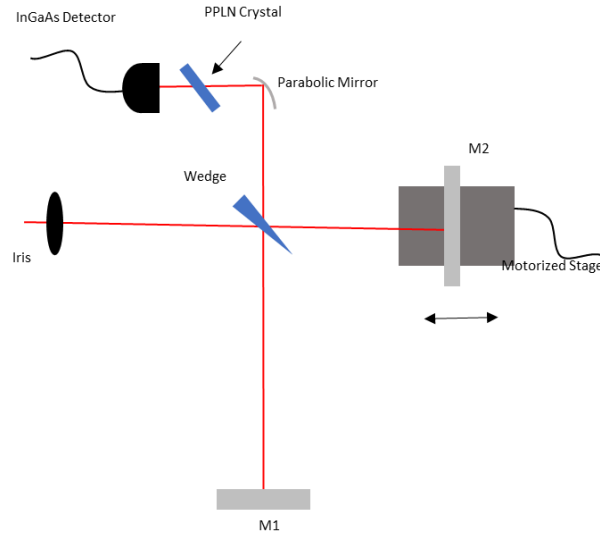
Figure 16: Schematic of non-linear crystal second order autocorrelator.

This autocorrelator is very similar to that used for the pump except for the addition of the PPLN crystal and the use of an InGaAs detector.

Investigation on the individual pulse durations of each lasing peak for an OPO had yet to be done so that was the goal of this experiment. By manually gathering the interferogram data I could individually create an interferogram for each peak and analyze them separately.
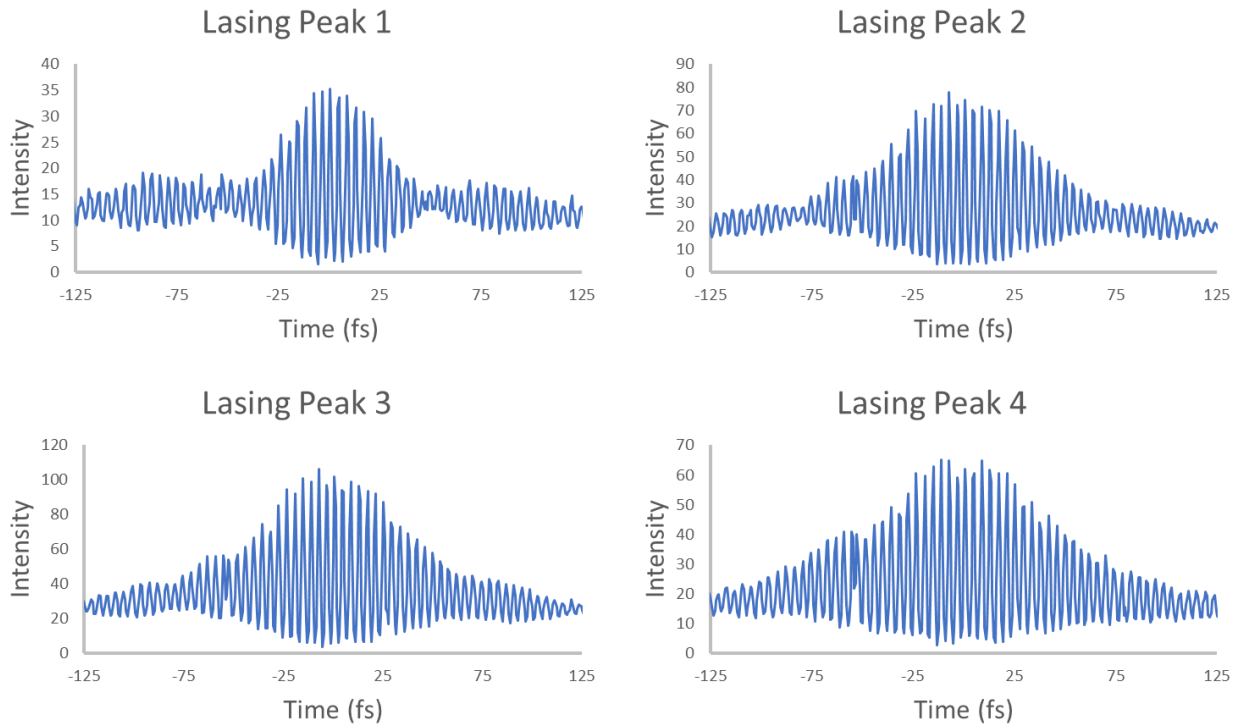
Peak	Pulse Duration
1	60fs
2	85fs
3	95fs
4	130fs

Figure 147: Individual lasing peak non-linear crystal autocorrelation interferograms and table of estimated pulse duration.

It was found that the individual lasing peaks did indeed have varying pulse durations and the shape even slightly varied. Pulse durations from 60fs, a completely degenerate peak, to 130fs, completely non-degenerate, were observed.

Chapter IV: Future Work

Since OPOs produces spectrums far into the mid-infrared with extremely high spectral resolution, this source is becoming very attractive for many applications. The system described in this thesis was designed and constructed for ultra-sensitive molecular detection applications. The future work of this system will be to rebuild it on a portable structure and begin lab experiments to determine is accuracy and usefulness in applications.

To accomplish this, a 2ft x 3ft optical breadboard that is 4.3" thick (for sufficient dampening) has been purchased to rebuild the GaP OPO on. This breadboard will be mounted to a custom aluminum structure with wheels for transportation via isolators (Isolators will allow for a more stable system like the one constructed in this thesis on a large air-ride optical table). All electrical components such as oscilloscopes, computer and stabilization components will also be on this cart. Additionally, this cart will have the detection scheme for the specific application. The idea of mounting the entire system and all its constituent components on a single mobile cart is analogous to that of medical equipment on medical carts at hospitals (The main application of this system is medical). By mobilizing the GaP OPO system we will be able to perform application testing at locations outside of our lab in CREOL.

Over 1000 different compounds have been identified to exist in human breath and many molecules have been correlated to specific diseases such as CO_2, CO, CH_4 and NH_3, which have links to medical diseases. While breath analysis is not currently used to officially diagnose such conditions, it provides an easy way to suggest that one should take further tests to diagnose. The reason laser breath analysis is exciting is because it is simple, accurate and completely non-invasive. To test our system for use in breath analysis, our group will be working with the Translational Research Institute for Metabolism and Diabetes at Florida Hospital and the Mayo

Clinic in Jacksonville Florida. Our colleagues at these facilities have been correlating high concentrations of certain molecules in the breath as markers for certain diseases and this system will provide a manner to accurately see many molecules across a broad spectral range at once.

For the second application of this system we will be working with Dr. Subith Vasu and Zachary Laparo of the Vasu Group in the Mechanical and Aerospace Engineering Department at the University of Central Florida. They are working on analyzing and improving engine combustion reactions. The conditions of a combustion reaction are simulated in a device called a shock tube. They are currently using narrow wavelength diode laser to analyze the concentration of select molecules over time that break down as the combustion reaction proceeds. In order to analyze all the molecules they are interested in, they would need more lasers than can be managed, and some at hard to attain wavelengths. Additionally, even with a significant number of lasers, they could not observe all the species at once. The use of our frequency comb system will enable us to analyze all molecules with a single source over a very broad spectrum.

These two applications will be an interesting investigation into the utilization of the unique mid-IR spectrum achieved from the system developed in this thesis.

Chapter V: Conclusion

The mid-Infrared spectral region is proving to be a key component to improving a multitude of applications, from bomb detection to monitoring CO_2 emission. To continue the advancement of these applications, a broadband coherent light source within the mid-infrared is needed. This thesis presented the development of a GaP OPO light source that can be implemented for the above applications, and more specifically breath and combustion engine analysis. This particular system has a high resolution coherent spectrum of over an octave wide, while still being efficient and only using a standard c-band laser. This system was made possible by the newly-developed highly non-linear, quasi phase matched, Gallium Phosphide crystal, suitable for telecom-laser pumping. With this compact system completed, research can now focus on the implementation of this device so that the mid-infrared region can meet its' full potential and change everyday life.

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