Variable Repetition Rate Ultra-Fast Ytterbium-Doped Fiber Laser for ARPES

By

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Thesis directed by Professor Dan Dessau

Abstract: Angle-Resolved Photoemission Spectroscopy, or ARPES, has been used since the early 1960s as a powerful tool to learn about the electronic structure of materials. To do this, ARPES utilizes the photoelectric effect to map out the momentum and energy bands of both bulk and surface electrons. ARPES requires high-energy photons, in the range of several eV to hundreds of eV per photon, to cause photoemission. This is generally done with synchrotrons; however, in the early 2000's ARPES was first done with a laser as the light source (Koralek & Dessau, 2007). Typically, and previously in our lab, this light source is a titanium sapphire laser but this comes with several shortcomings. To vastly improve this system, I have designed, built, and characterized a novel Ytterbium-doped fiber laser system that uses chirped pulse amplification to achieve an average power of ~90W with repetition rates from 20 kHz to 50 MHz and pulse energy of 900 μJ with pulses of ~300 femtoseconds. This is a significant improvement from typical Ti:sapphire in terms of power, pulse energy, repetition rate, and stability. These improvements offer new ways to upconvert from using BBO's to HHG due to the increase in peak intensity.

Acknowledgments

I would like to thank my committee, and especially my PI Dan Dessau for his support in this project. I would also like to give a big thank you to Bryan Berggren for his help on this project and Bryan, Nyah, Hope, and Shae for their help in the editing process (making this at all readable). Finally, thank you to my parents not only for supporting me academically but just generally putting up with me.

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Chapter 1

Introduction

Since the 1960s, Angle-Resolved Photo Emission Spectroscopy (ARPES) has been a powerful tool in studying materials (Borisenko, 2024). Both laser, synchrotron, and plasma ARPES are used to learn about the electronic properties of various materials, from organic superconductors to topological insulators. Using a plasma excited gas source for ARPES is done but is fairly uncommon and therefore we will focus largely on synchrotrons and lasers. ARPES is generally done with synchrotrons, which are capable of generating several hundred eV photons by accelerating electrons to near light speeds, and then using these electrons to create high-energy photons as accelerating charges produce electro-magnetic waves (Wilson, 1996). Synchrotrons are very effective and result in energy per photon that is essentially unattainable by lasers; however, it is not without its drawbacks. Since synchrotrons require accelerating electrons to near light speeds they are expensive and large. The average synchrotron is around one billion dollars to construct with upkeep cost around 100 million a year and generally a few kilometers in circumference (Wild, 2024). This means they are not common so beamtime is limited, and usually requires traveling. Laser ARPES however, is much more cost efficient, being a percentage of the cost of a synchrotron, and comes with its own benefits. Although we can not reach the same energy per photon we are able to get comparable resolution with the added benefit of coherent light. Since light is produced through accelerating charges at synchrotrons this is not possible, and; therefore, unique to laser ARPES. The most important benefit is the ability to use a process known as pump-probe. This involves exciting the electrons on a material then using high-energy photons to probe the surface, i.e. do ARPES. By changing the delay between the pump and probe pulses we can observe electrons as they decay from the excited to the ground state (Freericks, 2008). This gives access to a new domain of data, time resolved ARPES, and is impossible in synchrotron ARPES.

Chapter 2

Purpose and Background

2.1 Purpose

Ultimately, this laser's purpose is to do ARPES, and to effectively do ARPES we need high pulse energy, and high repetition rate. Pulse energy is the energy stored in a single pulse from the laser and is calculated by dividing the average power by the repetition rate. High pulse energy corresponds to peak intensity with the same pulse length and this is important because this laser will be used to drive nonlinear processes resulting in upconversion to higher energy photons. These processes' efficiency are related to the peak intensity; therefore, having higher pulse energy results in much more efficient up conversion later down the line. Therefore, there are two ways to achieve this; first, we can increase the average power. This is largely a function of the system design, although there are tweaks that can be made along the way to improve this without changing any major components. I will discuss both the components and these small tweaks that were made along the way throughout this thesis. Second, we can decrease the repetition rate. Although this increases the pulse energy, it also comes with some downsides; most notably a decrease in average power if the repetition rate gets too low. There is also a hard limit on the lowest repetition rate possible as we start over-pumping or the amplification medium can start self-pulsing. This is a result of over-pumping the rod and would cause significant damage to not only the final amplification stage but also any optic near the amplifier. Another factor we need to consider is stability of the laser. Poor stability either in power or mode will cause anomalies in ARPES measurements; therefore, must be avoided. The final consideration is compressing the ultrafast pulses as much as possible in order to maximize the peak intensity. Since we will be using nonlinear optics, such as Beta Barium Borate (BBO) or Lithium triborate (LBO), to convert the 1030 nm laser to 206 nm via 5th harmonic generation, we need incredibly high intensity to have efficient upconversion. In our case, this means peak intensity of ~5 trillion Watts. This laser will vastly improve pulse energy, stability, peak power, and repetition rate not only compared to our current Ti:Sapphire system but also in comparison to the standard for laser ARPES.

2.2 Background

Let's start by describing the old system and then characterizing it. Our Titanium-sapphire laser, as with all Ti:sapphire systems, consists of a Titanium-Doped Sapphire that is pumped with a 532 nm CW laser to create the desired population inversion. Then a seed pulse is sent through causing stimulated emission. Then through the use of mode locking, a tunable cavity, and an optical compressor, we can achieve ultrafast pulses, at around 40 fs. These pulses are around 800 nm (~1.55eV) and are amplified into powerful enough pulses for upconversion.

First, let's look at the power output and energy of each photon of our current laser system before and after upconversion. Our current system operates at around 810 nm before any upconversion. In contrast, we start with slightly lower energy per photon at 1030 nm; however, the major difference comes in power before any upconversion. The fiber laser outputs ~90 W of average power while the Ti:Sapphire laser is only at 12W of average power but the Ti:Sapphire is three times faster, therefore if operating at the same repetition rate we would expect around 3 times higher peak intensity from the fiber laser. With this much more efficient conversion can be achieved as upconversion is a nonlinear process. This generally involves combining two photons into a single photon although sometimes even more photons can be summed. This process will be discussed more in depth later, but it is important to note that the efficiency of this upconversion is dependent on the field strength squared. The more intense pulses will lead to much more efficient upconversion; therefore, resulting in more power after upconversion. The Ti:Sapphire laser takes 1 W average power of compressed 810 nm pulses and upconverts via a 4th harmonic BBO setup to get around 100 uW average power of 6 eV photons. Given the much higher pulse energy of our system, we can expect order of magnitude improvements to these final numbers for upconverted power alone.

Next, let's look at the stability. The main difference here between the two systems is the fact that the current system is fully a free space amplifier; therefore, small environmental changes can render the system incapacitated. The new system is done largely in fiber which means these same environmental changes: pressure, air currents, vibrations, or temperature, have almost no effect on the system. This results in the new laser being significantly more stable both over long and short timescales. The fiber laser's largest power fluctuations are around 0.2% in comparison to the old system which had fluctuations of several percent. Furthermore, these fluctuations were sometimes several percent, often causing the system to shut down. These power fluctuations lead to variations in mode quality which causes issues later in upconversion. This is a non-issue for the new system as two of three amplification stages and the oscillator are all in fiber, and therefore essentially immune to environmental changes meaning power fluctuation never causes a system shut off.

Finally, let's consider the repetition rate. The old system operates at two different repetition rates, either 20KHz or 50KHz. Changing between these two rates required significant realignment so it was not feasible to do often. In contrast, our current system can take any repetition rate from 20KHz to 50MHz and requires no realignment to do so and can be done via a digital input.

Overall it is clear that the new system is a substantial improvement to not only our current Ti:Sapphire system but also Ti:Sapphire systems in general. Since Ti:Sapphire systems are the standard for laser ARPES this represents an improvement for not only our lab but for the entire field. With higher power, pulse energy, stability, and significantly improved ease of use the new fiber system represents a significant step forward for our laser ARPES setup.

2.3 Chirped Pulse Amplification

Chirped pulse amplification is the basis for not only the fiber laser but also the titanium-sapphire laser. Ultrafast lasers have extremely high peak power which leads to inefficient amplifications and damage to optics. For example, if we attempted to send femtosecond pulses through our gain medium we would not only get very inefficient amplification, if the amplification stage even survives, but would likely damage optics as the electromagnetic flux would be too high. As exciting as it may sound to destroy some optics, it would not be helpful for ARPES, and would instead be a very expensive firework. To get around this, a process known as Chirped Pulse

Amplification (CPA) (Strickland) was invented by Donna Strickland and Gérard Mourou in 1985, for which they later won the 2018 Nobel Prize in Physics. CPA is a process of stretching out ultrafast pulses in time, then amplifying the stretched out pulse before finally recompressing the pulses. To stretch out the pulses we introduce chromatic dispersion to the light, meaning the shorter wavelength light ("bluer") is pulled back temporally and the longer wavelength light ("redder") is pushed forward.

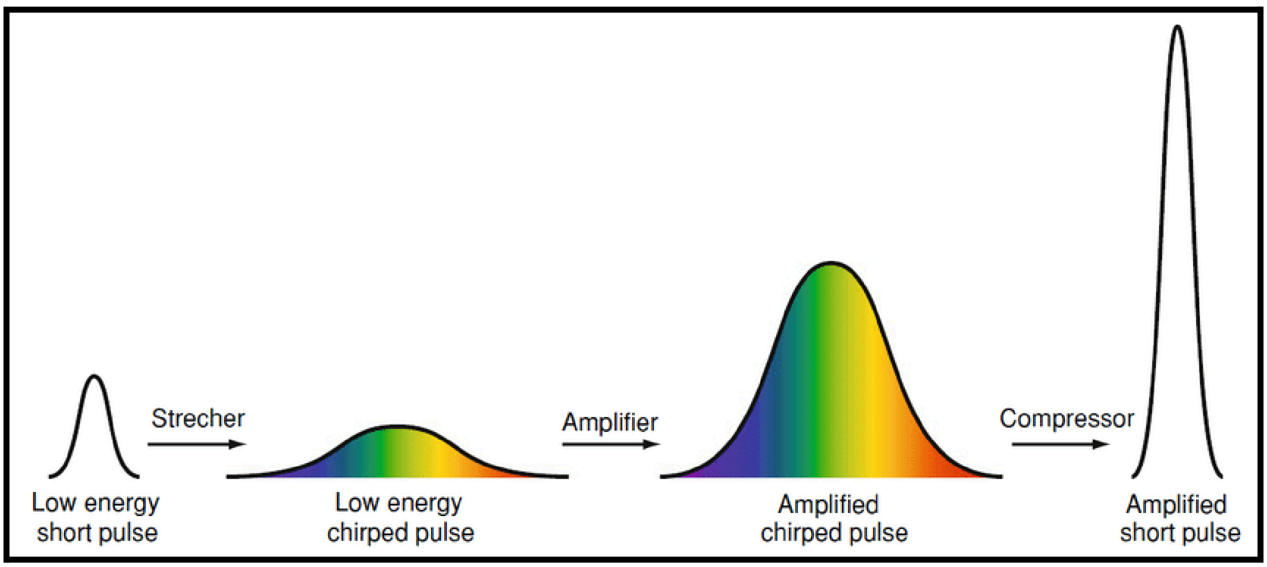

Figure 1: CPA. Start with a short pulse that is then stretched, amplified, and finally compressed. (Saxena, 2019)

To do this we take advantage of dispersive optics which alter light's path differently based on wavelength, like a prism, resulting in the redder and bluer light taking paths of different lengths; therefore, stretching the pulse in space and time. These dispersive elements, notable gratings, are now commercially available. Since the initial ultrafast pulses have a near Gaussian frequency distribution, this results in a new pulse that is significantly longer in time, with much lower peak power. In general this is 5 or 6 orders of magnitude increase in pulse length, and therefore in peak intensity. We have added frequency-dependent phase to the pulse and this is what stretches the pulse in time. With this lower intensity we can now amplify the pulse. This gives two major benefits: firstly, there is no fireworks show in the middle of the lab, secondly, we can achieve much more efficient amplification of the pulse as the amplification can occur for more time which allows for much higher peak powers later on. After we amplify the stretched pulse, the compressor removes the frequency dependent phase we introduced earlier in the system.

In a perfect world, the compressor could remove all of the phase introduced to the pulse in the stretcher, but in reality we end with a pulse that is noticeably longer, temporarily, than the incident pulse. In theory, we want to remove all of the dispersion as this would result in a pulse with a flat spectral phase. This means that all the frequencies of light present have the exact same phase and this would be the shortest possible pulse. This is limited by the Fourier transform limit and therefore is the theoretical max, but in reality this is impossible. In large part this is because every optic that interacts with the pulse adds some dispersion, although this is small compared to the stretcher, the randomness and amount of interactions makes compressing the pulse to the initial unstretched length effectively impossible. To remove this phase, we add frequency dependent phase in the other direction - if blue light took a longer path in the stretcher, now red light will take the longer path. By adjusting the position and angle of the gratings we are able to control the 2nd and 3rd order dispersion. This is not a perfect process and because of this the output pulses will be slightly longer due to 4th, and higher order, terms in the frequency dependent phase. We make a great effort to eliminate any 2nd or 3rd-order dispersion, but higher-order terms have negligible effect and for our purposes are not worth obsessing over. This process will be discussed much more in depth in regards to our laser later, but for now we have completed the general CPA process!

Chapter 3

System Overview

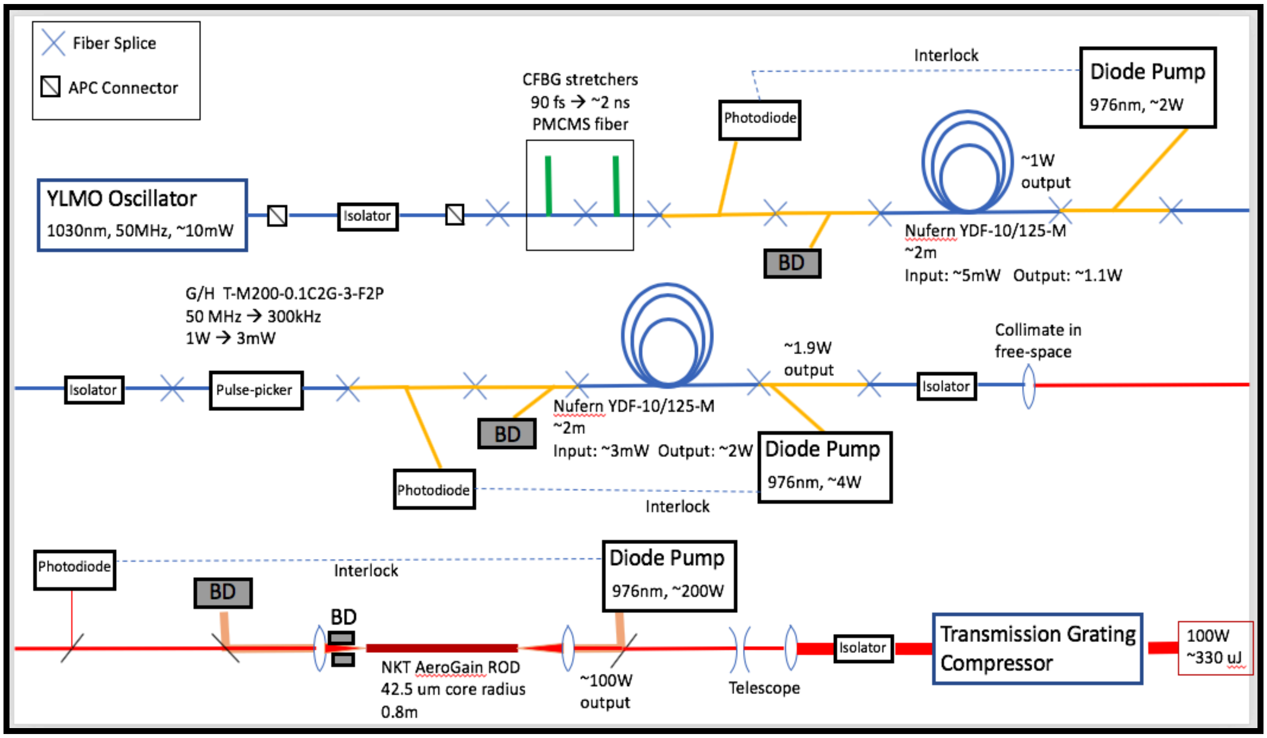

Figure 2: Schematic of the whole system with calculated values. (Berggren, 2023)

3.1 YLMO

Although the general process for CPA is universal there are some oddities in every CPA setup that are important to understand. In our laser we start with a YLMO oscillator (Menlo Systems, 2019) outputting 1030 nm light at 50 MHz at ~10mw with pulses of ~90 fs. This is very low power but we use it as a starting pulse so what is more important is the mode quality and the short pulse length. With a full width half max (FWHM) pulse length of 90 fs and a Gausian frequency distribution this oscillator is perfect as we can stretch out the pulse via chromatic dispersion and then recompress the pulse back into the femtosecond regime as the initial pulse was so quick.

3.2 Stretcher

After we have these ultrafast pulses they then go through a two-stage stretcher which takes the pulse from 90 fs to ~2 ns. To do this we take advantage of Bragg gratings in the fiber (Tosi, 2018). Bragg gratings work by having a penetration depth, the average depth light travels before being reflected, that is wavelength dependent. For example, “bluer” light could travel a short distance before getting reflected back while the “redder” light would travel a longer distance. This can be roughly imagined as a series of mirrors that only reflect a specific wavelength and transmit all other wavelengths where the reflected wavelength gets longer as you go down the series. With this in mind it is clear that the shorter wavelength, or bluer light, will travel a shorter distance before being reflected and therefore will end up spatially in front of the longer wavelength light as the redder light had to spend time traveling before being reflected. This simplified chirped fiber Bragg grating can be seen in figure 3. In reality, the design and construction of Bragg gratings is much more complicated, but this gives a good intuition to the processes at play. Through the use of two of these Bragg gratings we can take the 90 fs pulses from the YLMO, described above, and add chromatic dispersion to them resulting in ~2 ns pulses.

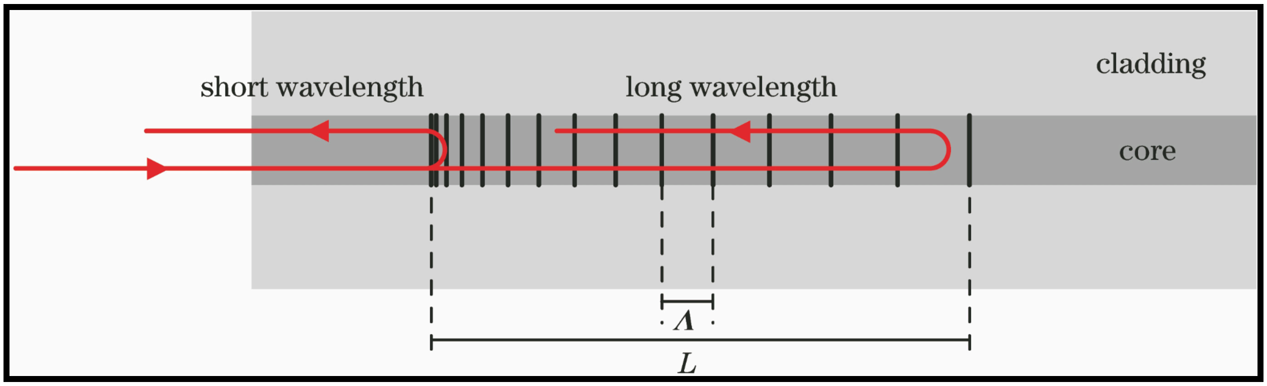

Figure 3: CHirped Fiber Bragg grating. (Xu & Li, 2022)

Although efficiency in this step is not vital, as the output is used to seed the first fiber amplifier, groups have reported efficiencies of ~95 percent using this type of Bragg grating (Paschotta, 2022). This is not something we measured in our system, but we can assume similar performance. After the stretchers, we are left with ~5 mW, about half our starting power, which is more than enough to begin amplification. This pulse then goes through a three-stage amplifier. This consists of two fiber amplifiers and one free-space amplifier.

3.3 Acousto-Optic Modulator

We use the AOM as a pulse picker, this means instead of having 50 million pulses every second can choose exactly how many pulses we have per second (Gooch and Housego). This is what we refer to as the repetition rate and by using the pulse AOM we are able to control the repetition rate. This is extremely useful; if we did not have this ability there would be so many pulses per second that the amplification per pulse would not be enough to reach the desired peak intensity. The amplification is not only limited in gain but also average power. This means if we send too many pulses through, the average power limit will come into play and each pulse will in turn get less amplification. Instead, we can lower the repetition rate so that each pulse can achieve the maximum gain possible to maximize peak intensity. As we will be upconverting the pulse via nonlinear processes the higher the peak intensity the more efficient the upconversion and therefore the ability to maximize gain per pulse is essential for our system.

Additionally, there are benefits to having a higher repetition rate. A great example of this in laser ARPES is that some materials decay even in ultrahigh vacuum and materials that do not decay still have surface degradation significant enough to affect ARPES data. Due to this, the amount of time ARPES can be performed is limited, and a higher repetition rate means more data can be taken resulting in lower error and more resolved data. This is also true for samples that do not have this issue as ARPES scans are just quicker if the sampling rate, or repetition rate, is higher. Even if the higher repetition rate is not a necessity it still saves time to have a higher repetition rate. Since different materials will have different requirements in terms of time before decay, required photon energy for photoemission, or photon flux for emission the best repetition rate can vary from one material to the next. For example, if a material lasts forever then we have days to run a single scan, so sacrificing some repetition rate to maximize pulse energy is helpful. This is especially true if we are using upconversion that requires large peak intensity such as high harmonic generation (HHG). Furthermore, the data can be better with lower repetition rate due to space charge effects. Conversely, if a material is going to decay very quickly we can increase the repetition rate to maximize the amount of data we can take before this decay occurs. As a result, the ability to change repetition rate is actually extremely useful for ARPES.

Now that we understand the purpose of the AOM, we can discuss how it actually acts as a pulse picker. To use the AOM as a pulse picker we use the Bragg diffraction. This is when materials have a different index of refraction when sound waves are passing through them. In our case, we use tellurium dioxide because of its ability to transmit 1030 nm light along with being very susceptible to Bragg diffraction, having a photoelastic coefficient of .14. Along with tellurium dioxide we use a piezoelectric to exploit this fact and cause Bragg diffraction. This is a material that vibrates at some known frequency when a current is run through it. This vibration creates sound waves, in our case at radio frequency, which can penetrate through the tellurium dioxide and cause Bragg diffraction. When the voltage, and therefore, acoustic waves, are on, the Bragg diffraction kicks the light out of its path. In this case, the AOM is acting as a closed shutter. However, when the voltage is turned off the light passing through is not diffracted and therefore can pass through unaffected; now the AOM is acting as an open shutter. The main advantage of this process in comparison to an actual mechanical shutter is the speed at which it can change from open to closed. A real shutter would have to physically move to either block or unblock the light which takes a significant amount of time. Some of the highest-end mechanical shutters on cameras max out at around 1/10000th of a second. Although, this is not a hard limit we are still very far from the required speed of a few nanoseconds. In the AOM on the other hand, the speed is limited by the time it takes for the acoustic wave to pass through the material, generally on the time scale of a couple nanoseconds. This means we can achieve significantly faster oscillations, by 4 or more orders of magnitude, with no fragile moving parts.

3.4 Fiber Amplifiers

After the pulse has been stretched out, it runs through two fiber amplifiers. The first of which takes our 5 mW incident pulse and outputs ~1 W. This is then run through a pulse picker which outputs around 3 mW which then goes into the second fiber amplifier, which amplifies the pulse up to ~3 W. These amplifiers are Ytterbium-doped fiber amplifiers (Coherent, 2020). The general ideas and processes are similar to those that are discussed in the AERO-Gain section, but we will quickly discuss the limitations and advantages. Since we are in fiber there are limitations to peak intensity as there are several nonlinear effects that could degrade the pulse if we let power get too high. These can range from slight distortions in the beam profile to destroying the fiber itself. In order to avoid this we are limited to only a few Watts in fiber, in our case 3-5 W. The main difference, however, is the simplicity and stability this adds to our system. Since the light is completely contained in the fiber it is nearly unaffected by changes in the environment. For example, changes in room air pressure causes instabilities in our current laser and generally the environment can cause issues from pointing instabilities, to dust on the optics decreasing efficiency. However, none of these things are issues and this contributes to the stability of the system while still allowing for amplification. Furthermore, we can take some small percent of the light after each stage and measure it which allows for us to not only monitor the system but also have automatic shut offs if the power on any stage drops too low. This protects the laser from damage, because leaving a pump diode on when there is no seed light to clear out the built up energy can lead to over-pumping. For us this is largely avoided because if power is lost to some stage the system can recognize this and immediately turn off every other stage. These interlocks are possible in free space; however, it generally leads to much more insertion loss, power loss by implementing the system as it requires a beam splitter. Since it is done in fiber and early in the amplification we lose next to nothing in terms of average power while protecting the laser for several possible issues and helps allow for us to reach the desired 3-5 W in seed power.

We now are at intensities that are difficult to contain in fiber as they cause nonlinear effects in the fiber core which will cause unwanted dispersion and can even possibly damage the fiber. To avoid this we couple the laser into free space for the rest of the amplification.

3.5 Free Space- aeroGAIN-ROD

In free space these pulses are used as seed light for the AERO-Gain rod (*AeroGAIN-ROD NKT*, 2016). This rod is back pumped using 200 W of CW 976 nm light to achieve population inversion. This involves running the CW laser through the AERO-Gain rod in reverse to the seed pulse.

Figure 4: aeroGAIN-ROD 2.1 (*NKT Photonics*)

This rod is known commercially as an AERO-Gain rod, but is really just a Ytterbium doped amplifier. It works in the same way as other amplifiers: the pump light, in this case 976 nm, excites the electrons of the amplification medium, in this case Ytterbium, to some excited state. Let's call this the E_3 state as it corresponds to the 3rd energy level. These electrons that are excited to this E_3 state then quickly spontaneously emitted in order to drop to some E_2 energy level. This E_2 energy level is necessarily lower energy than the 3rd energy level and furthermore this 3rd energy level is much shorter lived than the 2nd level. Subsequently, we get a population inversion in the rod and it is now prepared for lasing.

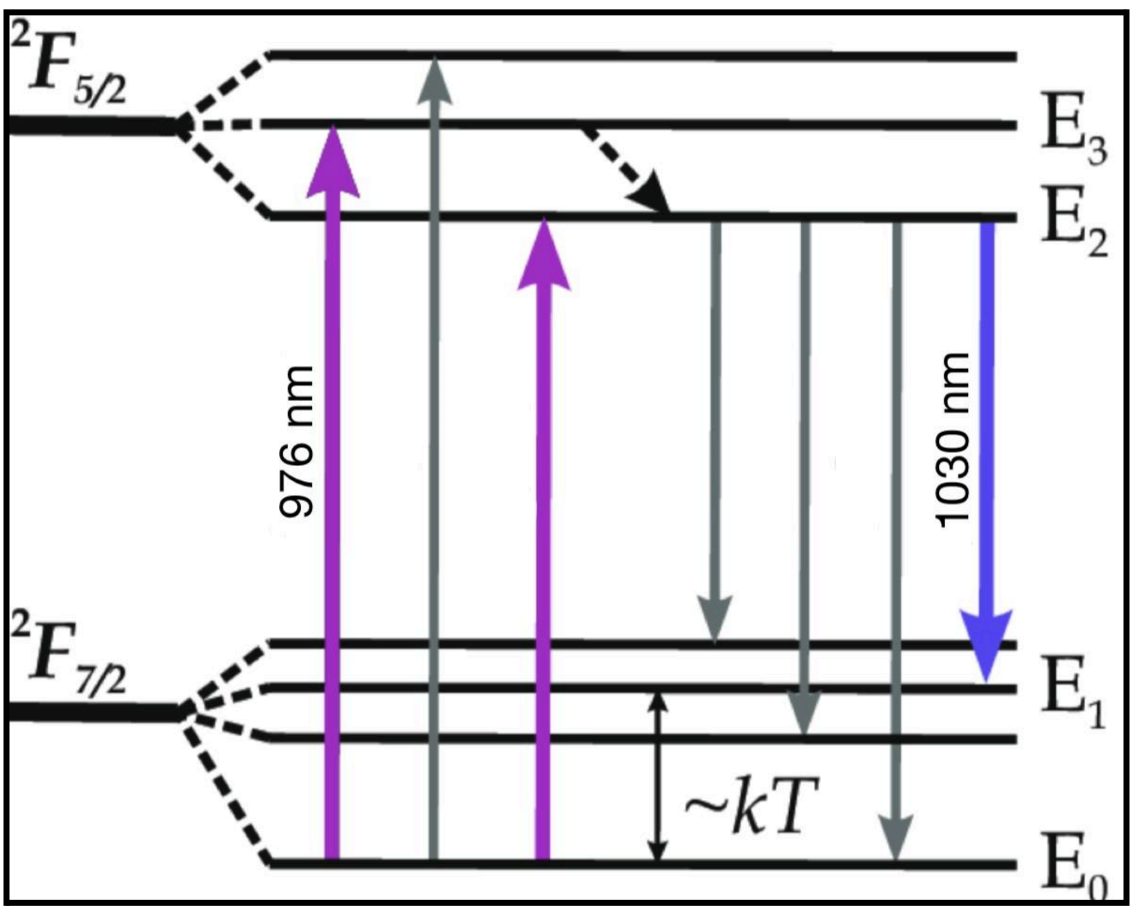

Figure 5: Energy levels of Ytterbium 2+ ions. We utilize the 968 nm excitation from E_0 to E_2 and then E_2 to E_1 for stimulated emission. (Novak, 2016)

This process to create population inversion takes some amount of time and it is why we can achieve higher amplification with lower repetition rate. The process is in some way analogous to charging your phone battery; for the first percent of charge it can charge very fast but then as it gets closer and closer to being full the rate of charging decays and more of the energy is lost to non charging. In our case, this is very similar as initially all of the Ytterbium sites are in their relaxed state and ready to accept a photon of 976 nm, then as the rod is “charged”, fewer of the Ytterbium ions are able to accept an incoming photon which leads to a decrease in the interaction cross section of the Ytterbium atoms. The cross sectional area represents the probability that an incoming pump photon will interact with a Ytterbium ion in the gain medium, as the interaction cross section decreases this probability decreases. As this area decreases, more photons pass through the rod without being absorbed and are lost as excess light so the gain is not just increased proportional to the time between pulses. This is also why pumping the laser with more light does not necessarily result in a proportional increase in pulse energy. Furthermore, we run into the risk of over-pumping the AERO-Gain rod. Overpumping can cause spontaneous emission, corresponding to 1030nm, that is amplified, and this can damage the rod. If spontaneous emission is amplified then there is no control over the direction of the pulse and this can lead to damage. Along with the lack of meaningful return to pumping harder over-pumping means there is a limit pumping.

Now that we have discussed this pumping process we can talk about the lasing process. We now have an electron in the 2nd energy level, and the gap from the 2nd to the 1st energy levels corresponds to a photon of 1030 nm. Now using a seed pulse of 1030 nm we can cause stimulated emission which amplifies the seed light and contributes to our output. Since this is stimulated emission the photons contributed to the seed pulse are perfect replicas of the incident photons. The photon resulting from stimulated emission is not only the same wavelength as the seed but is also in the same direction and with the same phase, we call this coherent. This process also largely preserves the cleaness, Gaussian profile both spatially and temporarily, of the seed pulse. We now have completed the final stage of amplification resulting in ~90 W of uncompressed pulsed light and are prepared to compress this pulse to achieve the maximum peak intensity.

3.6 Compressor

The process of compression is done with a dispersive element, something that reflects or transmits light of different wavelengths differently, and generally is done with reflective diffraction gratings. However, in our system we used Volume Phase Holographic (VPH) gratings (Wasatch Photonics), which are transmissive, but let's discuss the more commonly used reflective diffraction gratings. These gratings offer relatively high efficiency with compressors being built with reflective diffraction grating having 60-70 percent efficiency (Smirnov, 2015), but they are not without their downsides. Reflective gratings are made by tiny ridges on the surface, around the size of the wavelength of light, and these ridges are incredibly fragile. Conversely, reflective diffraction gratings are one of the easiest optics to damage and must be treated with immense caution. For my project this becomes relevant as our lab had a water leak which sprayed water over most of the free-space optics, including the gratings. Luckily for us, we used VPH gratings which are made by layering materials of high and low diffraction on top of another, and then sandwiching them between two optical windows. This means the grating is not a physical structure that can be easily damaged by water or any contact, and means this leak, although annoying, did not damage the compressor. The design also allows for contact to be safely made on the surface, making cleaning the VPH grating much easier than the reflective grating, where one is limited to using dry compressed air. VPH gratings also offer other benefits such as increased line density which leads to more efficient compression, in theory approaching 80 percent. Furthermore, VPH gratings can be made with much more consistency meaning much less scattering. With power levels as high as ours this is not only an efficiency concern but also a safety concern. Generally scattering in optical grating arises from inconsistencies in the surface forcing some light to be sent off in random directions, but in a VPH grating this issue is almost non-existent and, therefore, the main concern is reflections off of the substrate. Reflections can be remedied by applying anti-reflective coatings, and even though this is not perfect, these reflections are easy to predict and follow and, therefore, can be blocked without any safety concerns. When considering all of the benefits VPH adds it is clearly the best choice for compressing a high power system such as this fiber laser.

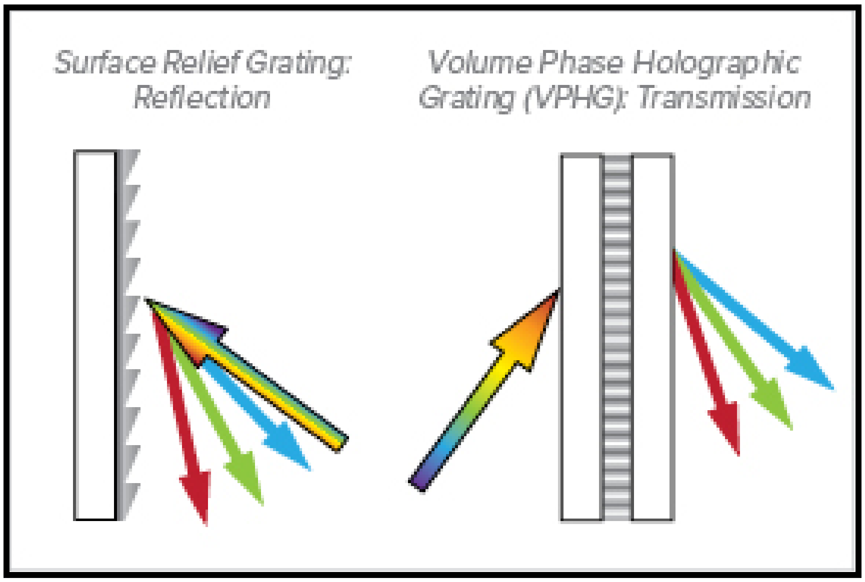

Figure 6: Light interacting with a reflective grating and a VPH grating. The darker material is the dispersive element while the white boxes represent substrate.

For our compression stage we have a double-pass compressor which removes the phase added by the stretcher, and all other optical elements, by sending the light through a compressor setup twice.

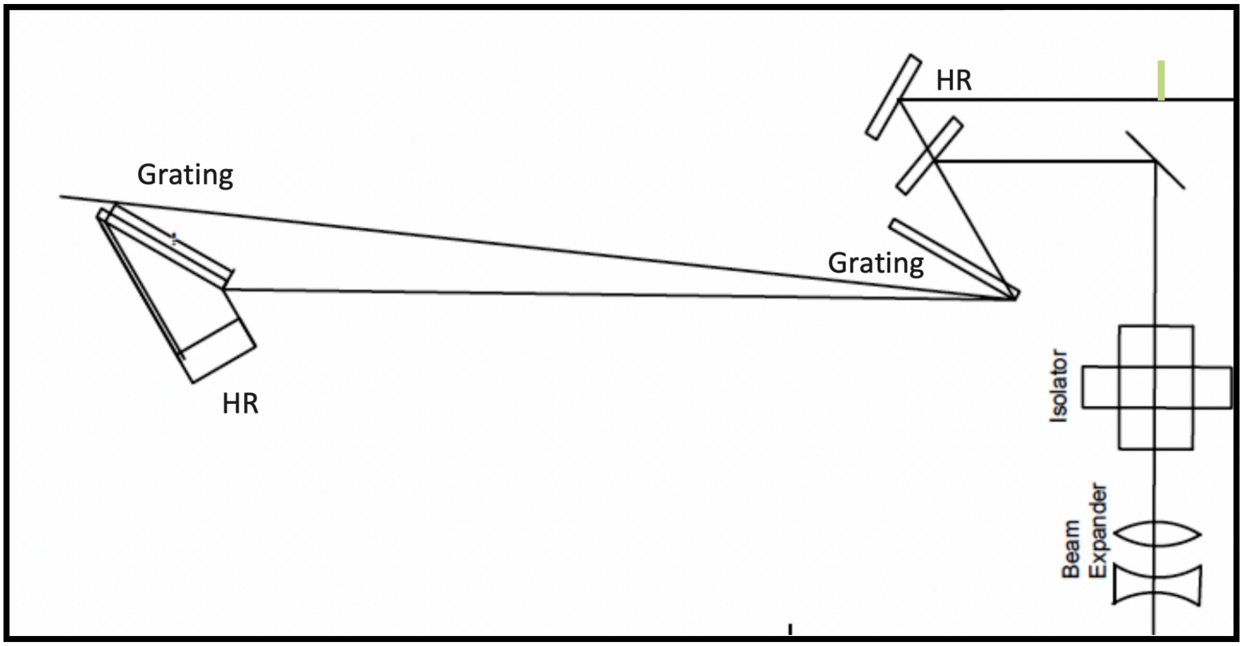

Figure 7: Double-pass compressor setup. Pulses go through the two gratings to the high reflector and back through the two gratings. After this a roof mirror sends the pulses through the whole system again before finally exiting. (Berggren, 2023)

This is not the most efficient possible setup as there are setups that can be slightly more efficient (Pedersen, 2024), i.e., less power is lost in reflections, but these setups take up much more space and, therefore, are unviable. Now after this compressor we finally have high-intensity pulses!

Chapter 4

Stability

4.1 Power Stability

In order to do ARPES we need both long and short-term power stability in our photon source. This has been an issue in our current setup. Free space lasers often have instabilities that cause power fluctuations, on both short and long-timescales. For our current system, without constant alignment optimizations these instabilities will become severe enough to cause a full loss of power. Because of this, overnight ARPES scans with this laser system are nearly impossible. Long-term instability is likely due to either pointing instability or the diode pumping the Ti:Sapphire going bad. Either way, there is not a simple fix to either the short-term or long-term instability, much less both. From this we can see the clear value of having a laser that is very stable in terms of power output.

Our former setup has power swings of several percent. On the other hand, the fiber laser has achieved power stability of .15W when operating at ~71W. This is a substantial improvement representing just a .2% change in power from the highs to the lows.

We still have a repeating pattern of power fluctuations that is on the 10 s of minutes time scale. This is due to the fact that the laser is water cooled and the water chiller which provides the cooling power operates in a 100% on or off state. The chiller will turn on, substantially cooling the AERO-Gain rod and all of the diodes, then they will slowly heat up until they hit some threshold temperature after which the chiller will turn on again. Since the gain profile of the AERO-Gain rod and the power output of the diodes depends on temperature these changes in temperature also cause changes in output power.

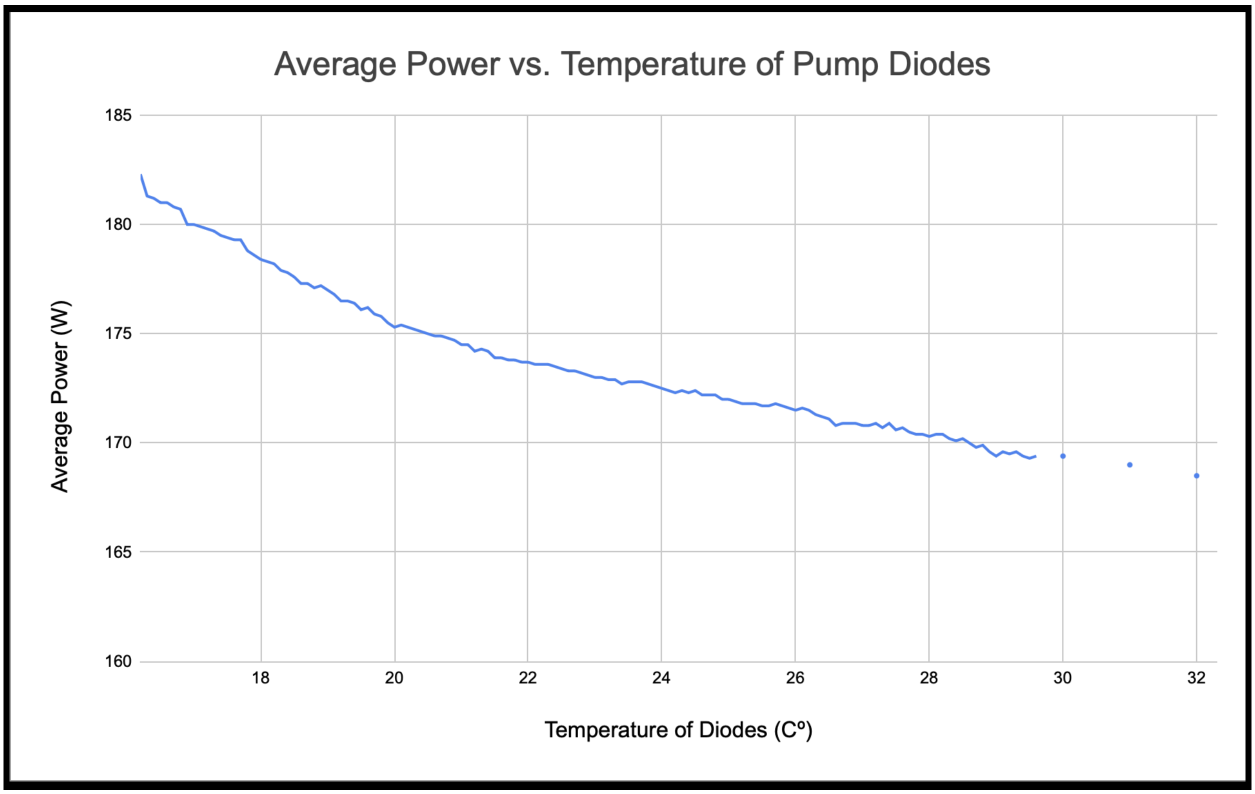

Figure 8: Graph of measured Power vs Temperature of Diodes. As the diode temperature increases the average power of the diodes decreases.

From this, it appears we could want the diodes to be as cool as possible as they output more power at these low temperatures; however, this is not the case. Since the temperature of the diodes and the AERO-Gain rod are inherently linked, as they are in the same cooling loop, it is difficult to determine the exact cause of decreases in output with lower temperatures. Since both the pump diode and the aeroGAIN-ROD are the same temperature the cause in these power changes can be narrowed down to two non-exclusive options. Firstly, the diode could have its center wavelength shifted due to the temperature changes, and this causes less efficient amplification in the AERO-Gain rod.

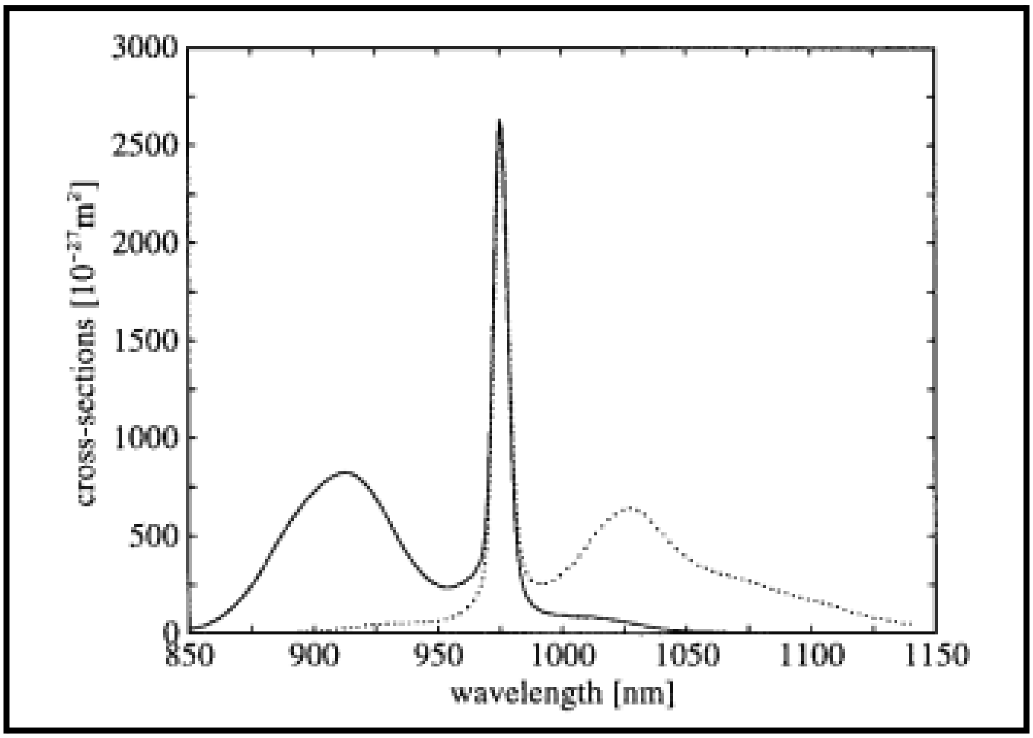

Figure 9: Calculated Cross-Sectional Area vs Pump Wavelength. The cross-sectional area shows a strong peak centered at ~976 nm. (Mukhopadhyay, 2010)

If the center wavelength were to be shifted by just a nm we would expect significantly less amplification. This option is definitely possible, but since the pump diode uses an optical cavity to determine its wavelength it is very unlikely that temperature changes would cause a significant enough shift in wavelength to have an effect. It is possible that the center wavelength does not shift, but that this extra power at low temperatures comes from a secondary peak. This actually was the case for our pump diodes and this means although the power outputted by the pump diode is increased the increase comes from another wavelength. This other peak was several nanometers off from 976 nm so it does not contribute significantly to pumping of the aeroGAIN-ROD.

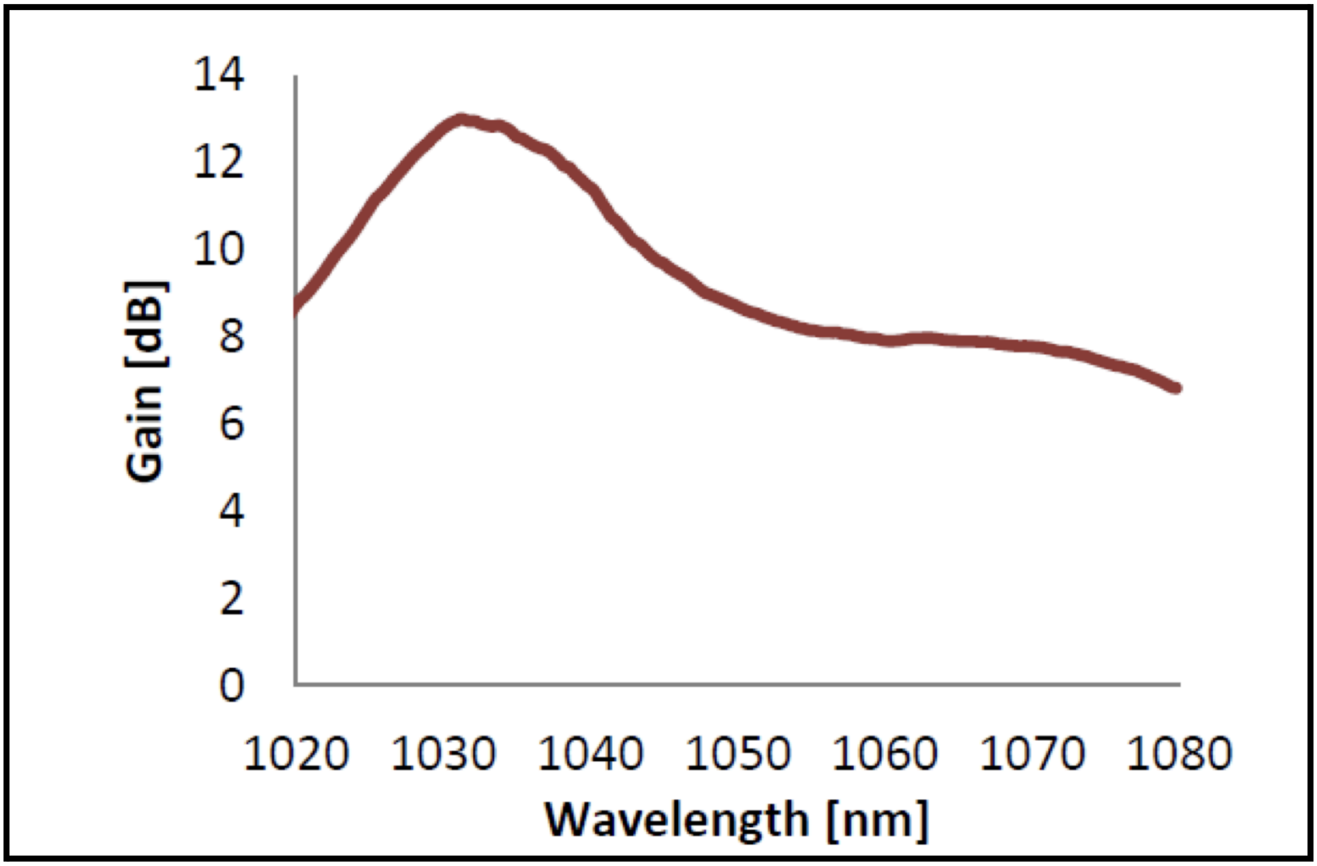

Figure 10: Calculated Gain vs Seed Wavelength per NKT's calculations. The gain shows a strong peak centered at ~1030 nm. (NKT Photonics, 2016)

Secondly, the AERO-Gain's gain profile is dependent on temperature. This is shown clearly when the rod, and diodes, are cooled to where the diodes are actually pumping harder and the amplified output still decreases. Even though this extra pumping is coming largely from a secondary peak which we expect to have only a small effect on pumping and, therefore, if the aeroGAIN-ROD amplification was not temperature dependent we would expect the power to stay about constant. This means we are giving the rod more power, or a similar amount of power at 976 nm, and it is still outputting less, meaning the amplification must be less efficient. We can conclude that the aeroGAIN-ROD's efficiency must be temperature dependent. This leaves us in a predicament. We want to keep the temperatures cooler, as this results in more power out of our diode with the same input, and it is better for the equipment. Running these Ytterbium-doped amplifiers at high temperatures causes increased photobleaching which can significantly decrease power output (Gebavi, 2012). Photobleaching is a process by which Ytterbium amplifiers can lose efficiency. This process occurs when photos cause Yb^{2+} to convert to Yb^{3+}, which has a different absorption profile, no longer peaking at 976nm. Because of this the efficiency of the amplifier decreases over time with usage at high power. Photobleaching appears to have a temperature dependence and is more common at higher temperatures so this means keeping the rod cooler has several benefits. Conversely, we also want to run the rod warmer so we do not get this shifted gain profile that causes a large decrease in efficiency so there is a limit to how cold we should run it; therefore, there is some ideal temperature at which these effects are balanced.

The first step of achieving this ideal temperature is to find at what temperature the output achieves a maximum. After several days of testing different temperatures for around an hour at a time it became clear that ~31^{o} C was ideal. When temperatures were slightly below we observed a slight decrease in power output until ~29.5^{o} C where a sharp decrease occurred. If temperatures got over 31 C there was a slow decrease in power. We want to keep the temperature above 29.5 C and as close to 31 C as possible. Initially to achieve this we set the setpoint of the chiller to 31 C. This meant if the laser got to 32 C it would kick on and cool everything to 31 C, as it has a one degree tolerance. Changing this massively improved our short term power fluctuation from 3% down to ~.3%.

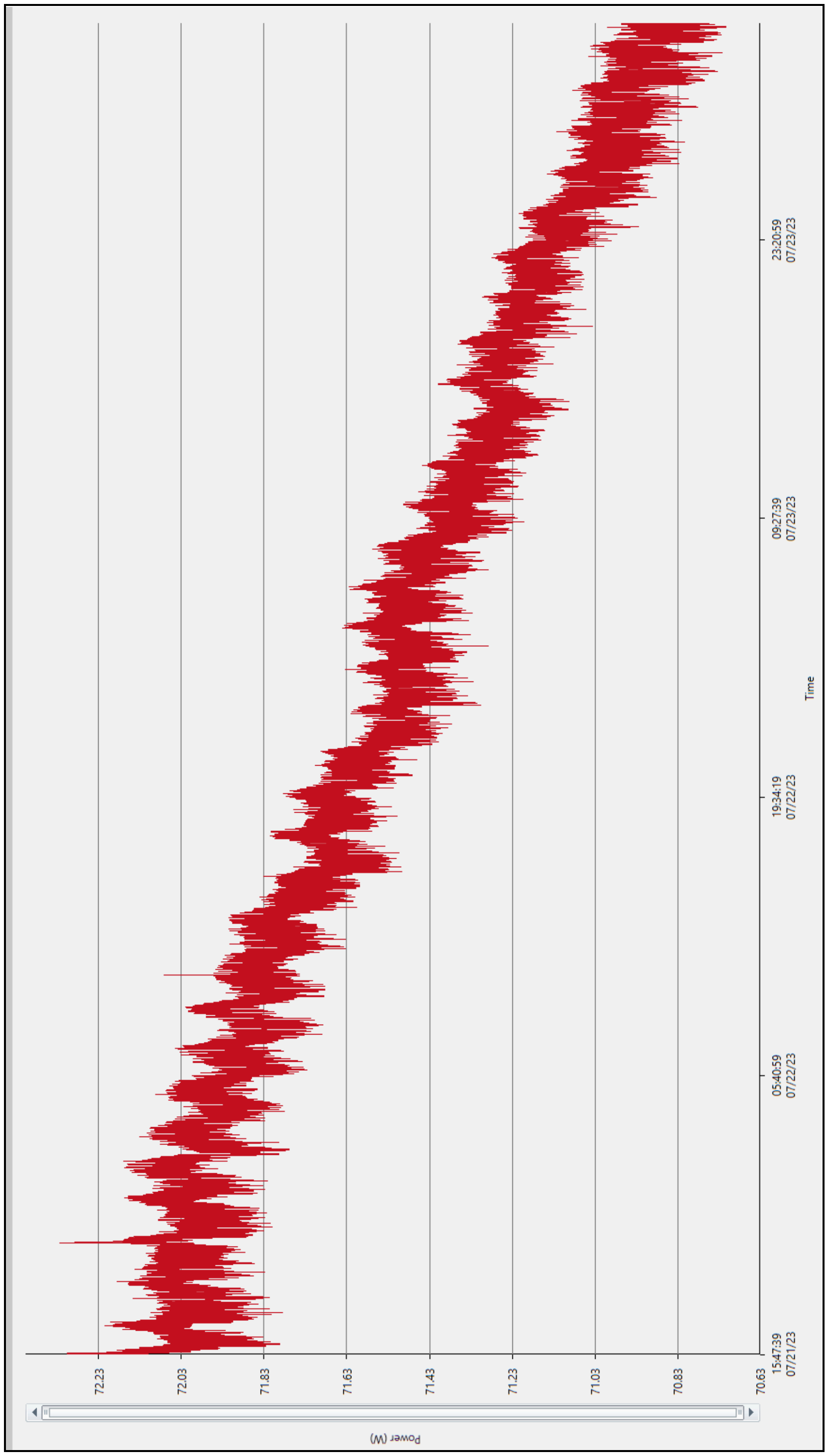

Figure 11: Power trend for the system over 66 hours. Short term fluctuations are around .2 W which represents fluctuations of around .25%. There is also a long term trend downwards in power that results in a decrease by 1 W.

To improve upon this further the setpoint was changed to 30.5 C such that the temperature would hover from below the highest output to above the highest output temperature. This resulted in, as predicted, the temperature fluctuation frequency being doubled while the amplitude of these fluctuations being halved. The change can be seen in the 1-hour scan below where the first half is the 31 C setpoint and the second is the 30.5 C setpoint.

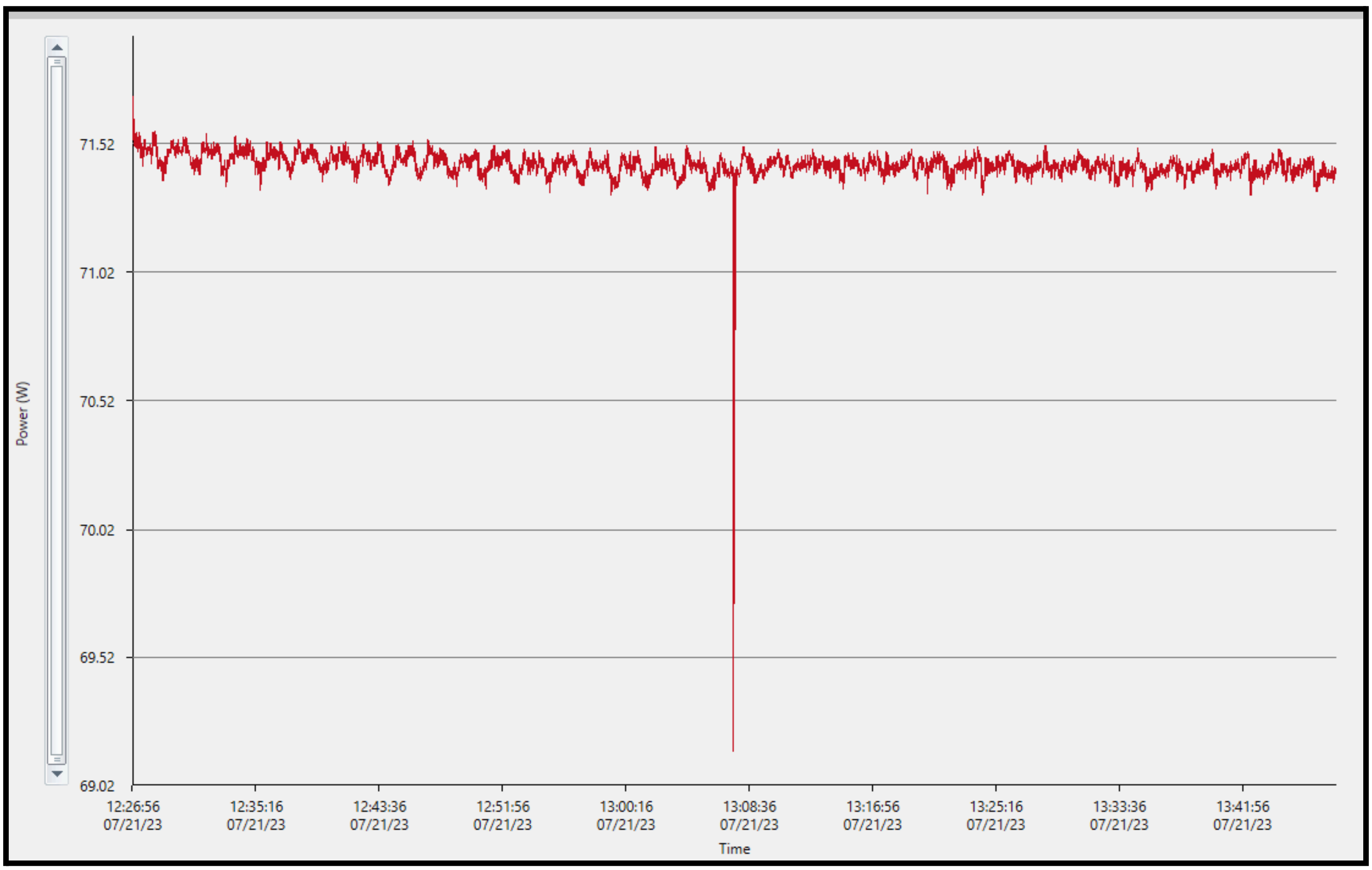

Figure 12: This plot shows a power trend over an hour. First 30 minutes the setpoint is 31 C. Second 30 minutes the setpoint is 30.5 C.

The second trend is visible in the 66 hour trend as there is a slight decrease in power over the trend. This change is not very large, but should be addressed. It is likely a thermal change, as we can rule out mode instability as shown in the section below. This change is likely due to thermal fluctuation in the lens that focuses the pump into the AERO-Gain. The lens focusing the pulse is protected by a beam dump, but we do not have this luxury in the pump lens as it is focusing the pump. This is not a large issue; however, because the seed light comes out at a smaller angle and, therefore, only passes through the glass of the pump lens which is AR coated for 1030 nm. Furthermore, we have the pump coming in at a much higher numerical aperture (NA) and, therefore, the pump lens is much closer to the aeroGAIN-ROD meaning the seed light does not have time to spread out and hit anything but the glass. If the signal coupling lens were being affected by thermal drift, we would expect to see the output mode change over time. We did not observe the mode shifting over time, so the instability is related to pump coupling. This lens is already aspherical and AR coated for 976 nm so the next step would be to heat the lens to a constant temperature. Then when the laser is off the heating would mean that when we turn it on the temperature would not change. Although this is feasible, the fluctuations in power are very small and this would add unnecessary cost and complexity to the system and, therefore, will be avoided. In the future if this slight instability is deemed too significant then this would be a good place to start, but for now we can ignore it.

4.2 Mode Stability

Having consistent power is not the only important measure. An optical spatial mode is the pattern produced by some 2D slice of a laser. In general you want the mode to be Gaussian, meaning symmetric and falling off as you go away from the center. The mode is important as it can affect amplification and upconversion efficiency, as these are determined by peak power, but even more important than the mode is the mode stability. Changing the mode means difference in field strength and pointing instabilities which can affect upconversion later in the system. To test the mode of the fiber laser we used near-field imaging to observe the mode directly over time so an understanding of near-field imaging is important. Since the core is only 85 micrometers, this is essentially a method of enlarging an image of the output beam right as it leaves the rod. To do this we run the output through a beam splitter, to not ruin the CMOS camera, then through another lens. Then by varying the ratio of the length between the first lens and the exit of the AERO-Gain rod, and between the second lens and the camera we can adjust the magnification. By placing the first lens close to the output, ~16mm, we can get an image of the light as it comes out of the rod. A simplified version can be seen below.

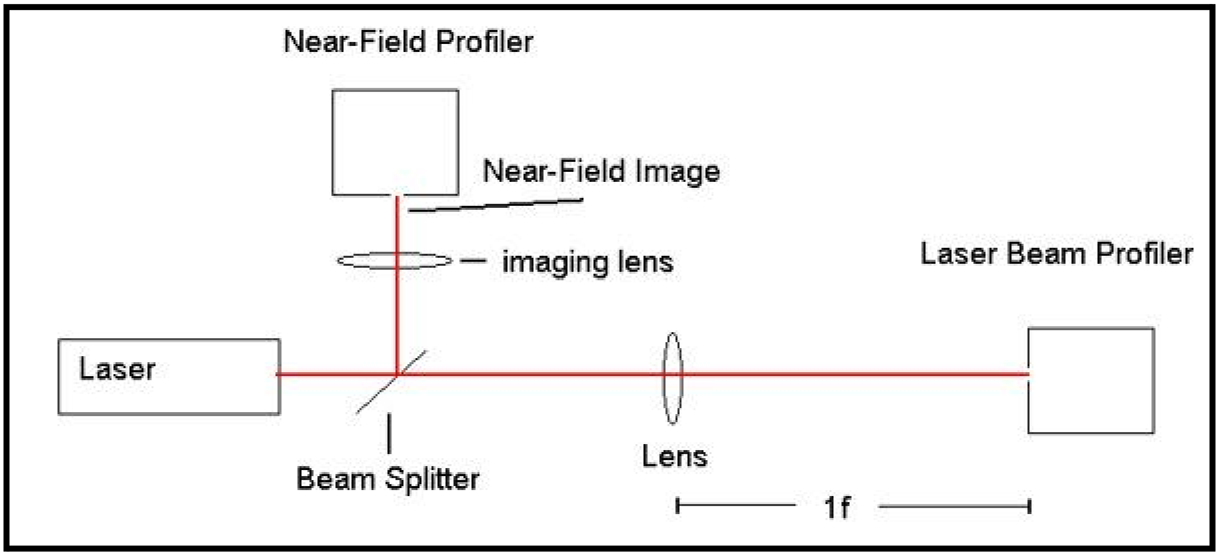

Figure 13: Near field imaging using a beam splitter and camera. (Cary, 2021)

Using this technique, we were able to compare the output mode right after turning the laser on and after a night of operation.

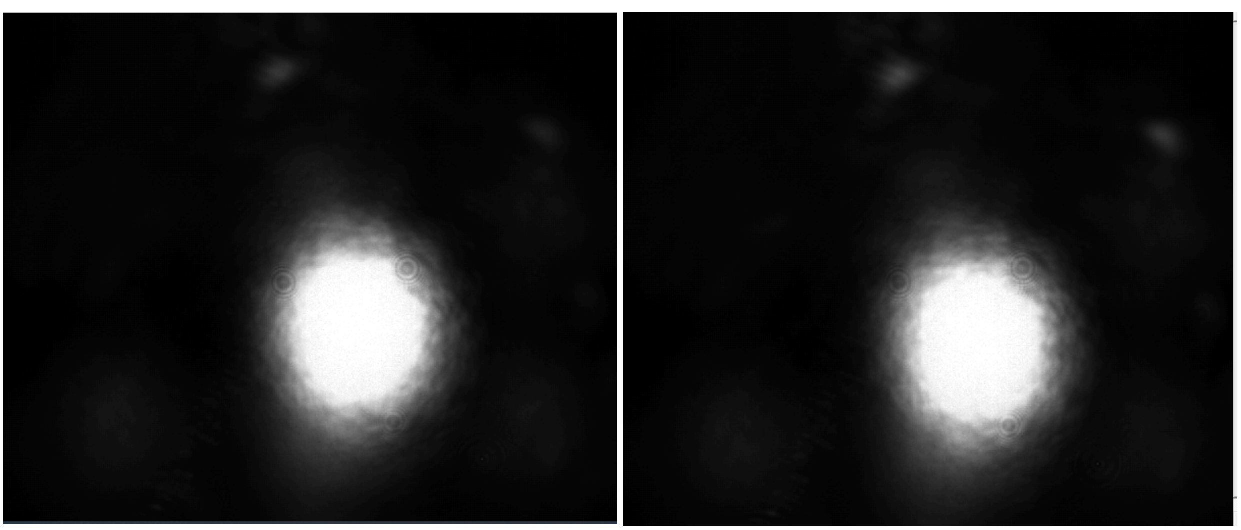

Figure 14: Two near field images of the amplified mode. The first image is immediately after turning the system on while the right is after 18 hours of being on at full power.

Unfortunately, this stability was not achieved on the first attempt. There were initially two main issues that were causing mode instability. First, the mount that the seed light lens was placed onto was settling, meaning that after adjusting it would slowly move for the first 24 or so hours. These changes were on the order of micrometers, but when the core of the AERO-Gain is 85 microns, this is significant. We were able to find this by checking the mode then turning the laser off and changing the mode again in the morning because then thermal drift could not have played a role. To fix this mode instability, we came up with a very intricate solution: buy a more expensive mount. This new mount was less susceptible to settling, which is when the position is changed slightly in the days after alignment, and therefore the decrease in power after alignment was minimized. Secondly, thermal fluctuations on the lens that focuses the seed pulses were causing the incoming light to change position slightly. To minimize these thermal fluctuations, we designed a water-cooled beam block with a pinhole such that the excess pump light would be blocked while the seed light was let through. With both of these solutions implemented, we were able to achieve near perfect mode stability over 18+ hour operations.

Chapter 5

Frequency Resolved Optical Gating

5.1 Background

Frequency Resolved Optical Grating, or FROG, was introduced in 1991 by Rick Trebino and Daniel Kane (Trebino & Kane, 1991) as the first way to fully characterize ultrafast laser pulses. Before this breakthrough it was largely thought that it just was not possible to fully characterize pulses in the femtosecond regime due to the fact that they were just so fast. Electronics max out in the 100's of GHz regime and, therefore, measurement of pulses this quick via electronics alone is impossible. Now, 30 years later, it is possible to take FROG measurements with a commercially available device and a graduate student can build a frog setup in a few weeks from scratch. This initial breakthrough, and following improvements, means that now fully characterizing a 300 femtosecond pulse is achievable by an undergraduate.

To characterize a pulse of light you need a few things. First you must know the intensity vs time of the pulse. This is fairly simple, and was measured for femtosecond pulses far before FROG, and is a relatively good measure of what a laser is outputting. However, if you are compressing a pulse, like in CPA, knowing the field magnitude and phase of a pulse gives valuable information about the pulse's compressibility. To measure this there are two main ways.

First, and most importantly as it is what we use, is multishot FROG. This process can be seen in figure 15 and consists of sending in several pulses into a beam splitter. After this pulse has been split into two identical pulses the pulses take similar length paths and then are overlapped in a nonlinear crystal, such as a Beta Barium Borate (BBO). This overlapping causes second harmonic generation, doubling the frequency of the light. Since this is a nonlinear effect the second harmonic generation is dependent on the intensity squared and; therefore, most of the doubled photons come from where the pulses have been overlapped. Furthermore, both of the identical pulses are sent in at some angle relative to the BBO, for example one pulse enters at +15 degrees while the other comes in from -15 degrees, and we place an iris to only allow photons exiting at 0 degrees to make it through. This means that if only one of these two pulses were to enter the crystal no doubled light will be detected as it will be blocked by an iris and, therefore, not be detected by the spectrometer. This means we are eliminating any photons generated by a single pulse as none of the light that is generated from a single pulse will enter the spectrometer. However, if a doubled photon is generated from both pulses, one photon from each identical pulse, then their angular momentum will be combined, resulting in a normally propagating output beam, and the light will exit straight into the spectrometer. Now the path length for one of the identical pulses can be varied by moving a mirror to extend or shorten the total path. By varying the path length and taking measurements of the doubled light's spectrum vs delay we now know the intensity vs time of the pulse based on how many doubled photons we detect. We also know the spectrum vs delay of each pulse, by measuring the spectrum of the doubled light. Instead of having to measure these values as fast as the light comes in, we can now measure them for each spatial part of the light by changing the delay between the identical pulses.

With this information it is possible to recover the phase. Although it is largely outside the scope of this project the general process that is used to retrieve the phase can be described briefly. We start with all possible -fields and limit down to which could satisfy the FROG trace. After this we know that the E-field must satisfy the mathematical form constraint, a mathematical requirement dependent on the non linear process. There is only one E-field that will satisfy both these constraints and therefore if we find an E-field that satisfies both we have finished. This is hard to do initially so to arrive at the solution we make a guess for the trace then take the closest point to the mathematical constraint and then from this mathematical constraint take the closest point to the trace. After several iterations of this process we can end at the correct E-field. Although this is a very simplified version of what is actually happening, it can provide some insight into how we are able to fully characterize a pulse with just the parameters described above.

5.2 FROG Setup

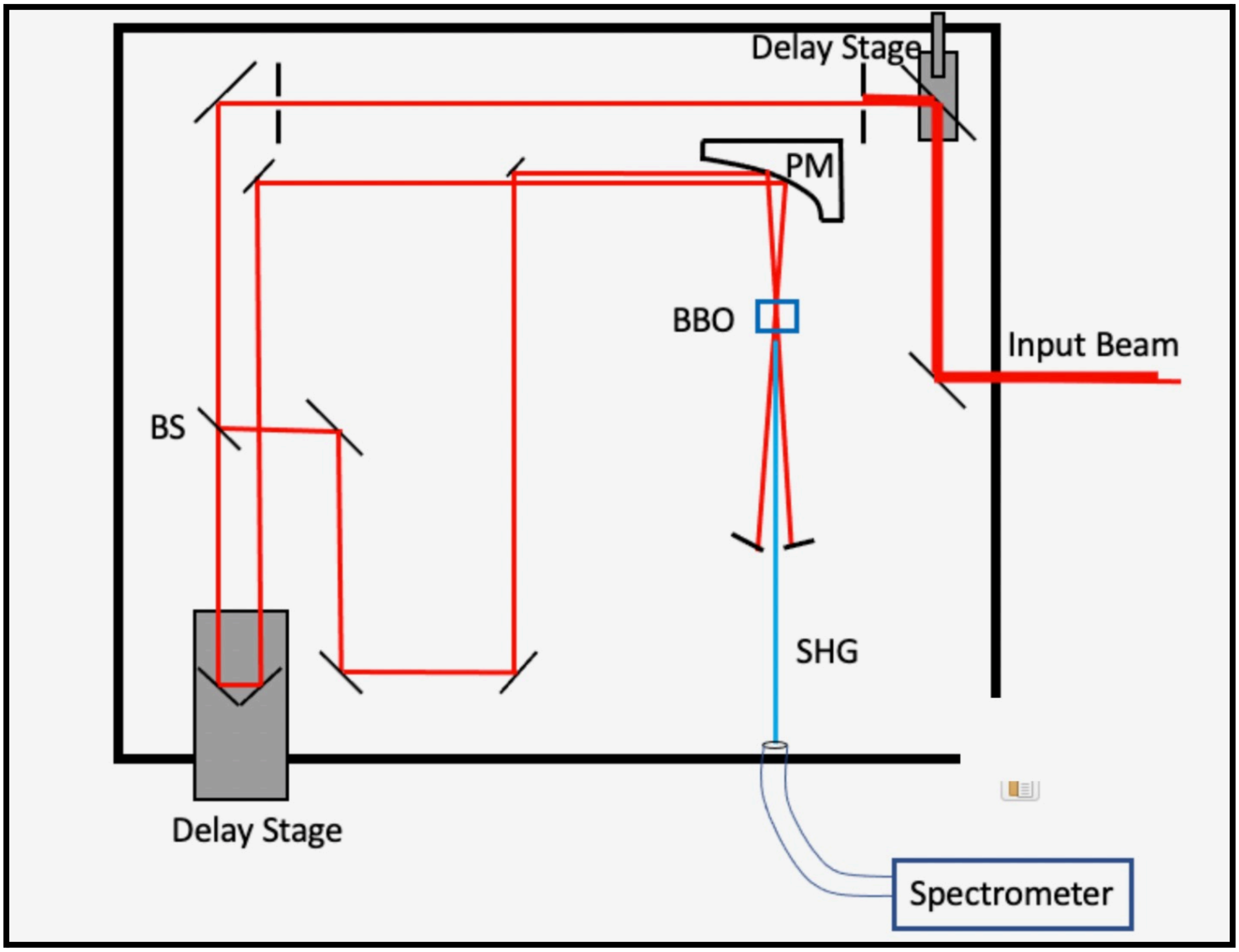

Figure 15: FROG setup. BS = Beam Splitter. PM = Parabolic Mirror. BBO = Beta Berrium Borate. SHG = Single Harmonic Generation. (Berggren, 2023)

Although our FROG setup is fairly standard there are a few differences. One important difference between our setup and most multi-shot FROG setups is the lack of any lenses in our setup. This is due to the fact that the pulse energy, and therefore intensity, is already high so there is no need to focus the beam in order to get second-order harmonic generation. Lenses also add dispersion that is not actually in the system. If the laser is optimized with a lens then we are not truly optimizing the system but rather the system after it has gone through the FROG. This also limits the risk of ionizing air in the compressor. Ionization could be dangerous not only for the optics but also the data as the electromagnetic field produced by the ionized air would affect the pulse itself. Other than these small differences our system can be thought of as any generic multi-shot FROG setup.

Chapter 6

Future Work

Although a lot of progress has been made, and the laser system is in a very impressive spot, there are still some future improvements that can be made to help implement the system into ARPES measurements. Firsty, the pulses must be upconverted to shorter wavelengths, and, therefore, higher energy. This can be done through the use of Lithium Triborate (LBO) or Beta Barium Borate (BBO). Based on the results of groups using lasers that are somewhat comparable it appears that LBO is more efficient for our wavelength and general power, or more importantly intensity. Before upconversion can occur FROG data must be taken in order to optimize the compressor. After upconversion the system will be ready for laser ARPES.

Even after the system is ready for ARPES there will still be some possible improvements. It is possible to couple the seed light into the final amplification stage through a monolithic fiber. This would mean less of the system is in free-space and therefore it would be even more resilient to environmental factors which would improve the power stability. Another way to improve the power stability would be to control the temperature of the pump diodes and the aeroGAIN-ROD with a proportional integral derivative (PID) temperature controller. As the short term power oscillations are due to temperature change, being able to control the temperature more precisely would decrease these fluctuations. A longer term goal would be to use the system to seed a high harmonic generation system which would be capable of producing photons with ~100 ev. This would put us in the realm of synchrotrons in terms of energy per photon in our own lab. Currently these HHG systems are very expensive and can not accommodate powers as high as this system produced but in the future this could be a monumental step.

Chapter 7

Conclusion

We have achieved 90 W average power with 900 μJ pulse energy before compression. This is more than enough for upconversion via BBO/LBO or HHG. Although there are still steps to be taken, namely upconversion, the key parts of the laser are not only set up but extremely successful. The system has power stability and mode stability that is far above the standard for ARPES, and this is due to the fact that the laser is mostly in fiber. This laser represents the next step for laser ARPES due to the increase in pulse energy, stability, repetition rate, and average power compared to standard Ti:Sapphire systems which allows for higher energy upconversion.

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Thank you