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Design, Construction, and Stabilization of an Adjustable Repetition Rate Frequency Comb for Precision Spectroscopy

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Abstract

Optical frequency combs have numerous applications across the sciences. One of the most powerful applications is in molecular spectroscopy, which takes advantage of both the coherence of lasers and combs' inherent broad bandwidth. One well-established design is the erbium fiber comb, which is popular due to its low cost and relative ease of construction. I have modified the traditional all-fiber design by introducing an adjustable free-space section which allows for adjustments to the path length, and thus the comb's repetition rate. This low-cost addition allows for repetition rate matching, a necessity for dual-comb spectroscopy, and active repetition rate stabilization, which is essential for long-term stability and precision measurement. Stabilization is achieved through passive and active means; active stabilization involves both temperature control and a piezoelectric transducer in the free-space section. Preliminary results, frequency comb spectra, and other diagnostic measurements are presented. This comb will be implemented in a dual-comb spectrometer to conduct rotationally-resolved measurements of small molecules such as hydrogen cyanide, and in a supersonic beam apparatus to measure larger molecules such as benzene.

Acknowledgments

This thesis was made possible thanks to several individuals. I would like to thank my initial readers, Professor Seifert and Professor Stewart, for giving me constructive feedback on my paper. I would also like to thank Connecticut College's Summer Science Research Institute for providing me with the opportunity to stay at the college over the summer to begin this research and spark my interest in lasers. I also give thanks to the sources of funding which helped cover the cost of spending the summer in the lab, ACS PRF grant #65007-UNI6, as well as the Yale-Taft fund for funding traveling to American Physical Society Conferences to present my work. Last but certainly not least I would like to thank my thesis advisor Professor Maser for providing me with the wonderful opportunity to work in his lab over the past years, for all the knowledge and skills he has imparted upon me, as well as all the help he has given me with classes and writing this thesis.

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Chapter 1

Introduction

1.1 Background

First developed in the late 1990s to count the cycles from atomic clocks , optical frequency combs are a Nobel Prize winning technology which has many applications in science and technology[1][2]. Optical frequency combs are made up of a series of equally spaced spectral lines each with a well defined frequency, making them a useful tool for spectroscopy, metrology, and communication systems. One of the primary applications of optical frequency combs is in high-precision frequency measurements. Optical frequency combs have revolutionized the field of optical metrology by enabling the development of extremely high precision optical atomic clocks, which are the most accurate time-keeping devices in the world. These clocks use the optical frequency comb as a reference to measure the frequency of light emitted by atoms, leading to a level of accuracy that surpasses traditional atomic clocks by several orders of magnitude [3]. Optical frequency combs are also used in the telecommunications industry for wavelength-division multiplexing (WDM) systems, which allow multiple signals to be transmitted simultaneously in a single optical fiber[4]. In addition to their uses in metrology, optical frequency combs have found many other applications in areas such as astronomy, molecular spectroscopy, and biomedical imaging[2]. Most importantly for our work is the use of optical frequency combs for molecular spectroscopy. The comb structure of the optical frequency comb enables the measurement of many molecular transitions with high accuracy and resolution[5]. Important applications of frequency comb spectroscopy have led to developments such as using frequency combs to detect methane leaks for use in oil fields [6] or even in medicinal applications such as developing a breathalyzer to detect trace molecular indicators of disease[7]. Their versatility and precision have made them a useful tool in many fields of science and technology, and ongoing research is occurring to explore new ways to harness their precision for even more applications.

1.2 Motivation

The ultimate goal of this research is to rotationally resolve the spectra of benzene using the technique of dual-comb spectroscopy. A vital step towards this goal includes the construction of a frequency comb with an adjustable repetition rate which can match the repetition rate of an existing frequency comb so that dual-comb spectroscopy can be performed[8]. I believe that spectroscopy of benzene will yield results that may be used in a wide variety of applications in fields of chemistry and physics. Information about the structure and behavior of benzene when absorbing frequencies of light could be used in industrial manufacturing settings where benzene is used in reactions to fabricate numerous materials such as plastics, resins, synthetic fibers, and lubricants[9]. In addition, with the extreme precision that can be achieved with optical frequency combs, there could be many uses of the experiment for modeling purposes. Although the spectra of benzene has already been vibrationally resolved near 1684 nm[10], we are hoping that we will be able to rotationally resolve the benzene spectra in the same near-infrared region in order to see each rotational transition in energy level and compare their frequencies to simulated results. Furthermore, differing theories about electron orbitals could be compared with the experimental results to make more accurate atomic and molecular models.

Chapter 2

Theory

2.1 Optical Frequency Combs

An optical frequency comb is a specialized laser with an optical spectrum consisting of a series of equidistant frequency lines. In the time domain, an optical frequency comb can be represented by a continuous train of light pulses, with each pulse emitted at a frequency dependent on the repetition rate of the frequency comb. In the frequency domain, the optical frequency comb can be represented by a series of lines at specific frequencies, resembling the teeth of a comb (hence the name frequency "comb"), similar to a combination of millions of single frequency lasers all in one as seen in Figure (2.1). Each comb tooth can be fully characterized by two frequencies, the repetition rate and the offset frequency, which can be put into a single equation

$$f_n = n f_{rep} + f_0 \tag{2.1}$$

where f_n is an optical frequency representing the nth integer comb tooth (where n is usually on the order of 10^6), f_{rep} is a radio frequency which represents the repetition rate (and spacing between the comb teeth), and f_0 is a radio frequency which represents some offset frequency from zero.

Equation (2.1) is quite useful as electronics cannot measure the fast oscillations of optical frequencies directly, but can measure the relatively slower oscillations of radio frequencies such as f_{rep} and f_0. The result is that previously impossible to measure optical frequencies are now able to be put in terms of measurable radio frequencies.

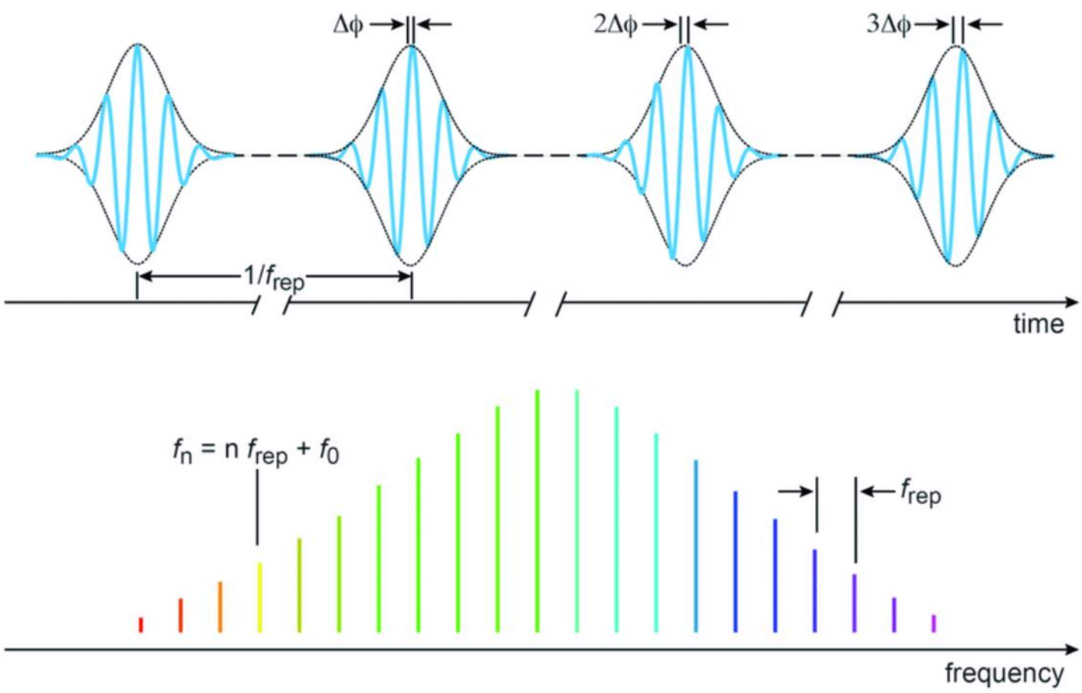

FIGURE 2.1: An optical frequency comb in the time domain (top) looks like a continuous train of pulses, the spacing of which is defined by the repetition rate of the laser. In the frequency domain (bottom), an optical frequency comb looks like a series of equidistant lines resembling the teeth of a comb (picture from Picqué and Hänsch 2019 [11]).

2.1.1 Mode Locking and Repetition Rate

Mode locking refers to a method of obtaining ultrashort pulses from lasers [12]. An erbium fiber laser will not generate an optical frequency comb until it is mode locked. Typically the pulse duration of an optical frequency generated by an erbium fiber laser can be on the order of 100 femtoseconds, or 10^{-13} s[13]. When a broad range of wavelengths of light are all present in a fiber, each wavelength oscillates at a different rate with shorter wavelengths oscillating faster than longer wavelengths as seen in Figure (2.2). For example, light waves at 300 nm (in the UV spectrum) will oscillate much faster than light waves at 700 nm (red light). As a result, each of these waves will be out of phase with each other most of the time in fiber. In a mode locked laser, the phase of each wave will line up at repeated intervals of time, which is called the repetition rate of the laser. In other words, the repetition rate of the optical frequency comb corresponds to how often these ultrashort pulses are created which will be the same as the spacing between the comb teeth.

In order to successfully mode lock the laser, it is necessary to modify the polarization state of the laser. The polarization state of light refers to the orientation of the electric field vector of light with respect to the fiber axis [14]. Polarization modulation, typically by bending, squeezing, or rotating a fiber, alters the polarization state of laser light as it travels through the fiber. Such polarization modulation also causes slight variations in the round-trip time of the light in the laser cavity, allowing for the frequencies to synchronise and create ultrashort pulses. All of the individual frequencies in the cavity are evenly spaced so that they can coherently form a pulse at certain intervals. For example, 1 Hz, 2 Hz, and 3 Hz waves would interfere at integer times of 1 second to form a pulse.

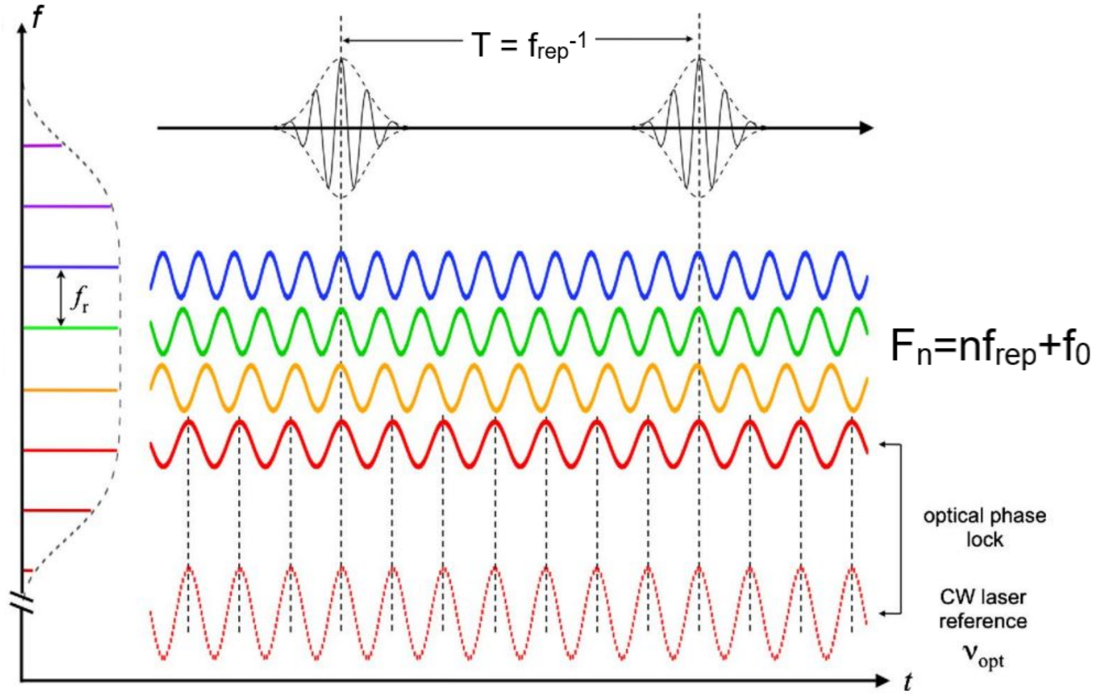

FIGURE 2.2: The oscillations of several different wavelengths of light (center) will periodically become in phase with each other at intervals of the repetition rate of the laser when it is mode locked, and create a discrete pulse of light (black, above). The Fourier transform of the pulse train provides the comb structure of the optical frequency comb (left) (picture from Diddams 2010[15]).

Fourier analysis plays a crucial role in the generation and analysis of optical frequency combs. The comb-like structure of the frequency spectrum arises from the Fourier transform of the time-domain pulse train that generates the comb. By taking the Fourier transform of the pulse train, the frequency components of the comb can be separated and analyzed individually. In other words, because the Fourier transform of a sine wave is a delta function at the specific frequency of the sine wave, the sum of sine waves produces a “comb” of delta functions. This allows for the precise measurement of the frequency and phase of each component, which is critical for many applications in metrology and spectroscopy[15]. Fourier analysis is also used in the detection and analysis of optical frequency combs. The spectrum of an optical frequency comb can be measured using a

Fourier transform spectrometer, which separates the different frequencies of the comb and provides a high-resolution spectral analysis[16].

2.2 Spectroscopy

Spectroscopy is generally referred to as the study of absorption and emission of light and other radiation by matter, but can also be broadened to include the study of the interactions between particles such as electrons or protons[17]. Each element on the periodic table has a unique set of wavelengths which will absorb light and excite an electron to a higher energy level[18]. Conversely, an electron can lose energy and drop to a lower energy level, emitting photons of light at those same wavelengths. In molecules, the principle is similar, molecules can absorb certain wavelengths of light related to electron excitement, but for molecules, absorption can also induce rotations and vibrations in the molecule. Measuring absorption wavelengths as well as rotational and vibrational modes can yield valuable information about molecules such as molecular structure, concentrations, electron configurations, and information about chemical bonds[19].

2.2.1 Spectroscopy using an Optical Frequency Comb

By using an optical frequency comb as a reference, molecular spectra can be measured over a broad spectral range with extremely high precision, making it possible to detect even weak spectral features that might be missed by other techniques. This has led to significant advancements in the study of chemical reactions and the characterization of complex molecules[5].

For simple absorption spectroscopy, a single optical frequency comb in combination with a spectrometer is sufficient, usually referred to as direct frequency comb spectroscopy (DFCS)[20]. One can simply pass the light from an optical frequency comb through a sample of gas and compare the the original spectrum with the spectrum produced through the sample. The absorption of certain wavelengths of light will be seen by a lack of those wavelengths in the spectrum that was sent through the sample of gas which had previously been present in the original comb spectrum as can be seen as an example in Figure (2.3). DFCS can also measure ro-vibrational transitions[21], but for these purposes we will be using dual-comb spectroscopy.

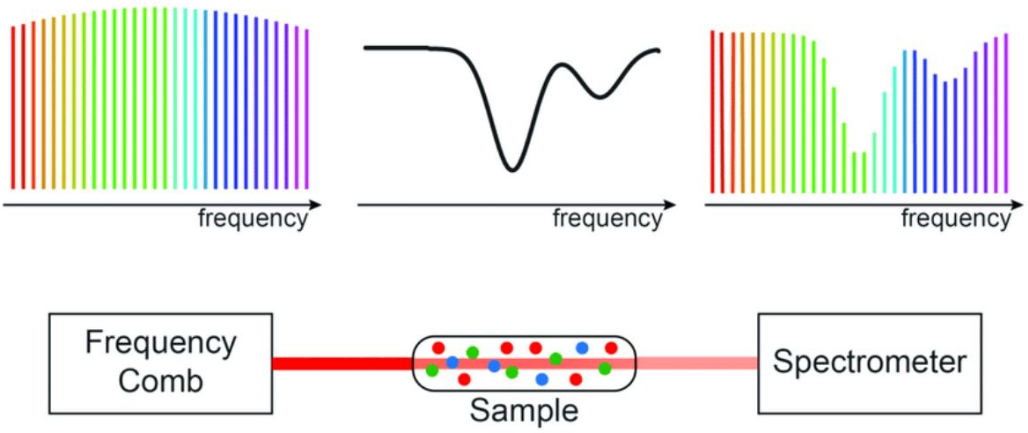

FIGURE 2.3: The light from a frequency comb can be sent directly through a sample to be collected by a spectrometer, any troughs in the spectrum indicate absorption of light at those wavelengths by the sample (picture from Picqué and Hänsch 2019 [11]).

2.2.2 Dual-Comb Spectroscopy

In order to obtain more information about a sample, such as rotational and vibrational modes, dual-comb spectroscopy will provide the necessary means. In dual comb spectroscopy, two combs are slightly offset in repetition rate by some Δf_r, usually within 1 kHz[8], so the spacing between the teeth in one comb is not exactly the same as the spacing in the other comb. When light from the two combs are combined, the individual teeth from one comb interfere with the corresponding teeth from the other comb. The interference between the two combs creates a radio frequency comb composed of beat notes that can be detected using a photodetector[22] as can be seen in Figure (2.4). The beat note frequency is equal to the difference between the frequencies of the two combs, which can be seen with the red and blue arrows in (a.) of Figure (2.4). If the dual-comb light is sent through a sample, the amplitude and phase of the beat note carry information about the sample's properties, which can be extracted using signal processing techniques. The precise spacing and phase relationship between the comb teeth are critical for achieving high spectral resolution in dual comb spectroscopy, so that the beat note created by the interference can be analyzed with high resolution and accuracy.

Dual-comb spectroscopy offers several advantages over direct frequency comb spectroscopy for ro-vibrational spectroscopy. Most importantly, dual-comb spectroscopy can achieve higher spectral resolution than direct frequency comb spectroscopy. In direct frequency comb spectroscopy, the spectral resolution is determined by the free spectral range of the spectrometer, which is the range of wavelengths or frequencies over which

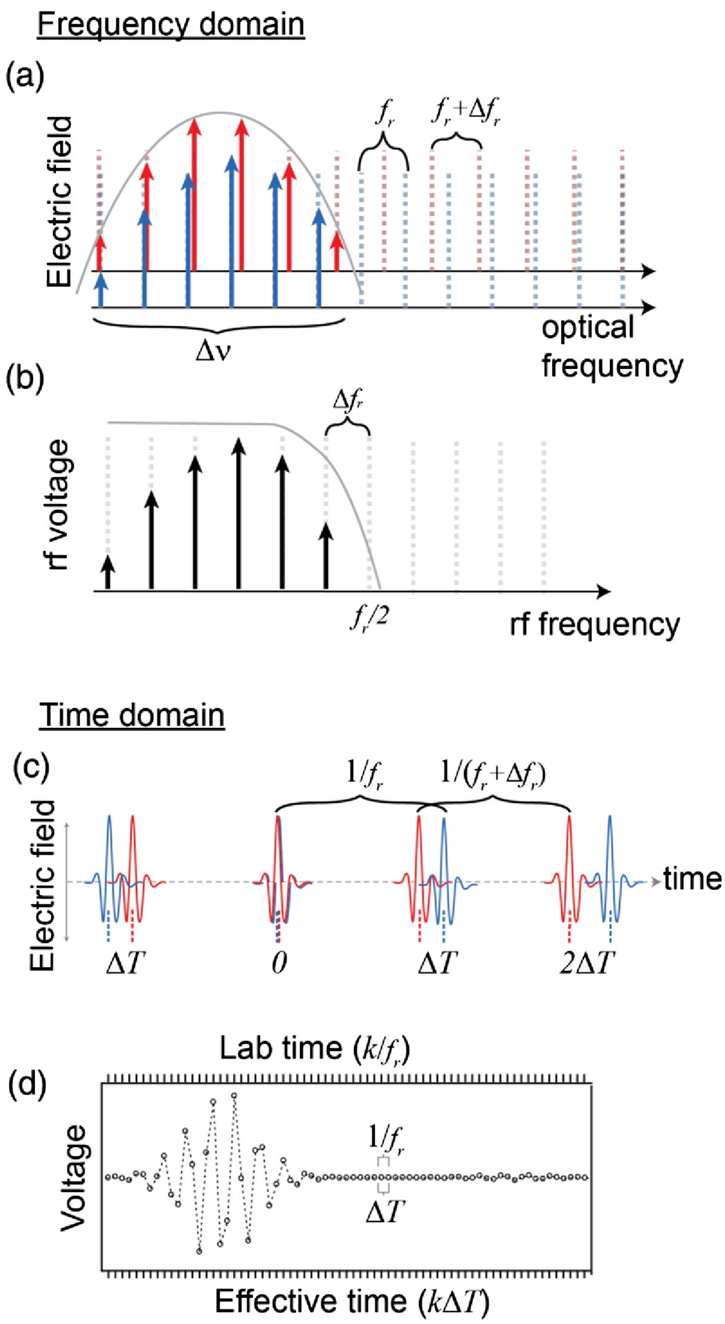

FIGURE 2.4: In the frequency domain, two optical frequency combs (a) (red and blue) are mixed to produce a (b) radio frequency comb. Equivalently looking at the time domain, the light pulses from each frequency comb will overlap at intervals of the repetition rate + Δf_r (c) to create an interferogram (d) with relevant spectral and absorption information about a sample, where k samples are taken in intervals of ΔT(picture from Coddington et. al. 2016[8]).

the spectrometer can operate without any overlapping of adjacent spectral lines as determined by the physical properties of the spectrometer, such as the spacing of its optical elements like gratings or mirrors. Because the comb tooth spacing is finer than the resolution of most spectrometers, if several comb teeth lie within the same transmission or reflection peak, they cannot be distinguished from each other by the spectrometer, limiting the resolution and potentially hiding important spectral information[8][23]. In contrast, the use of dual-comb spectroscopy enables much higher resolution due to the interference between combs creating the radio frequency comb with a significantly higher bandwidth, allowing for each individual comb tooth to be resolved. Dual comb spectroscopy can also provide much faster acquisition time of data than DFCS. In direct frequency comb spectroscopy, the high resolution of the spectrometer is achieved through mechanical scanning, usually of some diffraction grating to vary the angle of incidence of the light and cause the different wavelengths to be diffracted at different angles so a detector can be used to measure the intensity of the diffracted light at each wavelength. Such a process can be quite time consuming with higher precision requiring slower mechanical scanning. Dual-comb spectroscopy does not require mechanical scanning and can acquire spectra in a fraction of the time required for direct frequency comb spectroscopy[8].

Chapter 3

Experimental Design

3.1 Optical Frequency Comb Designs

3.1.1 Design Principles of Frequency Combs

Erbium fiber lasers are a well established method of optical frequency comb generation, with numerous successful designs [13, 24–35]. The frequency comb developed in this experiment is an adaptation of several base designs, which can be seen in Figure (3.1), modified to include broad bandwidth tunability of repetition rate.

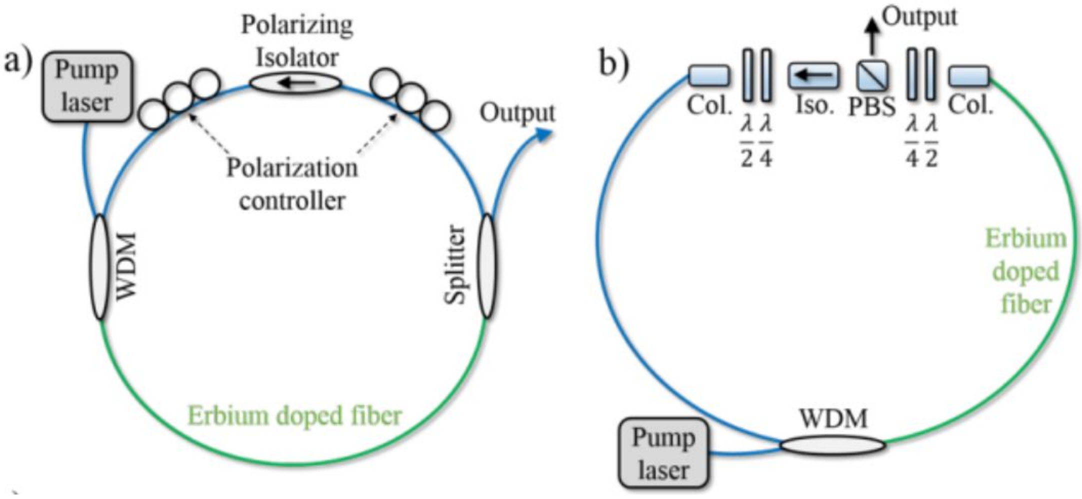

FIGURE 3.1: The base comb designs that were be used, with the base designs from existing literature by S. Droste [35]. Each design contains a WDM, Erbium doped fiber, and some form of polarization control using a series of $\frac{\lambda}{2}$ and $\frac{\lambda}{4}$waveplates or polarization controllers.

Commonly seen components of the erbium fiber laser, as well as their primary function, are as follows:

- **Erbium Doped Fiber (EDF)**: Erbium doped fiber is a type of optical fiber in which erbium has been incoporated into the fiber structure. Erbium doped fiber has a strong absorption band at 980 nm and an emission spectrum centered at 1550 nm. In erbium fiber lasers, erbium doped fiber is commonly pumped with 980 nm light from an external pump laser and the subsequent emission of light in the 1550 nm range provides light for the generation of a frequency comb in that same range.

- **Wavelength Division Multiplexer (WDM)**: A WDM is a fiber optic component that enables the transmission of multiple optical signals of different wavelengths in a single fiber [4]. In erbium fiber lasers, a WDM allows for 980 nm light to be pumped into the erbium doped fiber while simultaneously allowing the emmitted 1550 nm light to pass.

- **Isolator**: An isolator is a device which will only allow light to travel in a single direction. In erbium fiber lasers, an isolator is necessary to reduce noise and protect the laser from signals which may cause instabilities and shifts.

- **Splitter**: A splitter enables an optical signal to be distributed into two different fibers. In erbium fiber lasers, a splitter is commonly used to divert a percentage of the signal away from the laser in order to monitor the activity of the laser. Use of a splitter with other measurement devices can yield useful information about pulse repetition rate, optical frequency, and laser power.

- **Polarizing Beam Splitter (PBS)**: Polarizing beam splitters enables an incoming beam of light to be split into two orthogonal linear polarizations [36]. The use of a polarizing beam splitter is necessary in erbium fiber lasers to ensure that the output light of the frequency comb is polarized along the same axis.

- **Polarization Controller**: Polarization controllers are used to alter the polarization state of light through fiber. Polarization controllers can be used to bend, rotate, or squeeze fibers in such a way to achieve any arbitrary polarization state. In erbium fiber lasers, polarization controllers are necessary to achieve a mode-locked state and generate an optical frequency comb.

- **Collimator**: A collimator is a device with a lens which is able to focus a beam of light into a narrow path aligned in a certain direction. In erbium fiber lasers, collimators are very useful for aligning light out of fiber through a free-space section with a collimator on a receiving end of the free-space section to collect light back into fiber. Collimators for use in fiber optics will generally be connected to fibers by using a patch cord, which comes in two main variations: angled physical contact (APC) and physical contact (PC). APC connectors have a slight angle to the end of the fiber, which can help prevent light from back reflecting into the fiber, while PC connectors have a flat face and may cause light to reflect back into the fiber.

- **Circulator**: A circulator is a device with at least three ports which has been designed such that light entering from one port will exit through the following port [37]. For example, in a three-port circulator, light entering into port 1 will exit through port 2, and any light entering port 2, including reflected light, will exit port 3, not port 1. In an erbium fiber laser, a circulator is useful for separating light traveling opposite directions in a fiber.

Several designs of optical frequency combs were proposed and attempts were made to build each design to analyze viability in the experiment. The base design of the erbium fiber optical frequency combs as seen in Figure (3.1) are already well established, but alterations needed to be made to be able to adjust the repetition rate of the frequency comb. Thus, all of our designs incorporate some form of free-space section into the traditionally all-fiber frequency combs.

In order to achieve a repetition rate of 100.038 MHz, the same repetition rate as a previously built comb which will be used in conjunction with the newly designed comb, the lengths of the fiber and free-space section of the new frequency comb must correspond to the same repetition rate of 100.038 MHz. The repetition rate of the frequency comb is related to the length of the fiber by the equation:

$$f_{rep} = \frac{c}{n_{fiber}L_{fiber} + n_{air}L_{air}} \tag{3.1}$$

where f_{rep} is the repetition rate of the laser (100.038×10^6 Hz), c is the speed of light in a vacuum (3×10^8 m/s), L_{fiber} is the length of fiber in the comb, L_{air} is the length of the free-space section, n_{fiber} is the refractive index of the fiber which is 1.4682 at 1550 nm for SMF-28[38], the most commonly used fiber in our design, and n_{air} is the refractive index of open air which is approximately 1. If the free-space section is set to the center of its range at 5.5 cm, this leaves approximately 200.5 cm allocated to fiber in order to achieve the desired 100.038 MHz repetition rate.

3.1.2 Circulator and Mirror Design

The first attempt to create an adjustable repetition rate frequency comb involved the use of a circulator in the cavity connected to a free-space and adjustable mirror setup. The comb light would travel around the cavity and enter the circulator clockwise from the side of the splitter, where the light would then exit the cavity into an adjustable free-space section connected with a mirror. The light would then reflect off the mirror to be collected back into the circulator and continue clockwise throughout the cavity as seen in Figure (3.2). The distance between the free-space and mirror can be adjusted coarsely by moving a FiberPort Collimator forward or backwards along the connecting rail with the mirror, and more fine adjustments can be made with the micrometer knob and piezoelectric transducer.

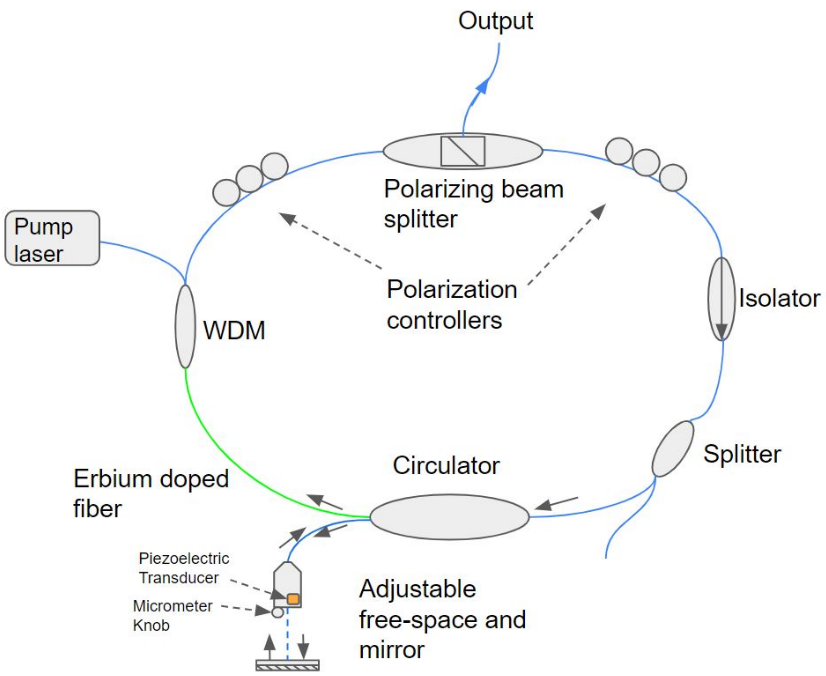

FIGURE 3.2: Light travels clockwise into the circulator out into free-space where it reflects off a mirror to be collected back into fiber where the circulator will send the light clockwise through the cavity once again.

The Circulator and Mirror design was discarded due to failure to achieve a mode-locked state. One issue that arose was the insertion loss of the circulator of approximately 1 dB, leading to a significant reduction in power in the cavity. Another factor which may have led to failure to achieve mode-locking was the use of a PC connector instead of an APC connector. Due to constraints on the length of the fiber, it is not feasible to splice an APC connector to the circulator and it was necessary manually melt and polish a PC connector to the fiber as PC connectors were the only melt-able connectors which were available in the lab. The use of PC connectors introduced the problem of light reflecting directly back into the fiber, introducing issues with transmission and alignment, which ultimately required a different approach to be taken to design the frequency comb.

3.1.3 Dual FiberPort Design

Subsequent frequency comb designs involve the use of two FiberPorts spaced a short distance apart mounted on rails which allowed for the ports to be moved closer or more distant as needed. The removal of the circulator and mirror setup slightly lessened the burden of constraints on fiber length of the main cavity and APC connectors had room to be spliced onto the fiber and used in the FiberPorts. Light would travel clockwise around the cavity and enter a collimator and travel through free-space to be collected by the opposite collimator where the light could then continue clockwise through the fiber as seen in Figure (3.3).

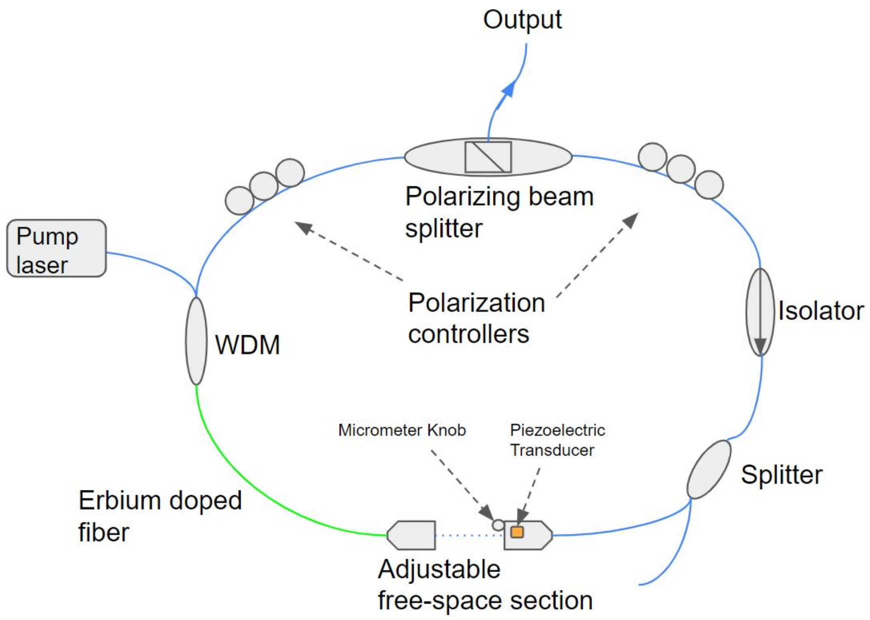

FIGURE 3.3: The adjustable free-space section features two FiberPorts which can be moved closer or farther for coarse length adjustments. A Micrometer knob and piezo-electric transducer is also present for fine adjustment of length.

The Dual FiberPort design was able to achieve a mode-locked state. The main difficulty that seemed to be present with dual FiberPorts was difficulty aligning light to minimize any power lost in the frequency comb. It was often quite hard to discern if the alignment was optimal, with one of the only reliable benchmarks being whether mode-locking was able to be achieved.

This design of an adjustable repetition rate frequency comb seemed to be promising enough to merit keeping the dual FiberPort setup, but several other factors required refinement to increase viability. Although the change from the circulator and mirror design allowed more fiber length to be allocated towards the main cavity, constraints on the length still caused significant trouble. The presence of the four main components as well as the two FiberPorts required at least one splice on either side of each fiber component, with each splice also requiring a rigid splice protector leading to reduced flexibility of the physical comb, causing issues when the comb was moved into a box for protection. On several occasions fibers in the comb broke, which required entire components to have to be replaced and re-spliced into the cavity, which motivated a refinement to the design.

3.1.4 Consolidated Dual FiberPort Design

The current design of optical frequency comb which has been implemented in the lab is similar to the previous design and uses the same dual FiberPort design, but the WDM, isolator, and splitter components have all been consolidated into a single combination WDM/Isolator/Splitter. Light is pumped into the cavity in the counterclockwise direction as before, but now the isolator blocks light in the clockwise direction, indicating that forward pumping is now being utilised as seen in Figure (3.4) as opposed to backward pumping which was used in the previous two designs in which the light was pumped counterclockwise into the cavity but the isolator blocks light in the counterclockwise direction.

This consolidation of components greatly reduced the amount of splices present in the comb and solved issues of fiber breakages due to lack of flexibility of the physical comb. The alignment of the dual FiberPorts still proved to be difficult but was able to be aligned well enough to allow for mode-locking to be achieved. With a design of adjustable repetition rate frequency comb which could be successfully mode-locked, further testing was able to be performed.

3.2 Stabilization of Frequency Comb Repetition Rate

Several factors will affect the stabilization and repetition rate of an optical frequency comb. Some key factors include [39]:

- **Temperature variations**: The stability of the laser cavity is affected by temperature changes. Most notably, the temperature can affect the length of the fiber with higher temperatures causing the fiber to expand and lengthen (decreases repetition rate) and cooler temperatures causing the fiber to contract and shorten (increases repetition rate).

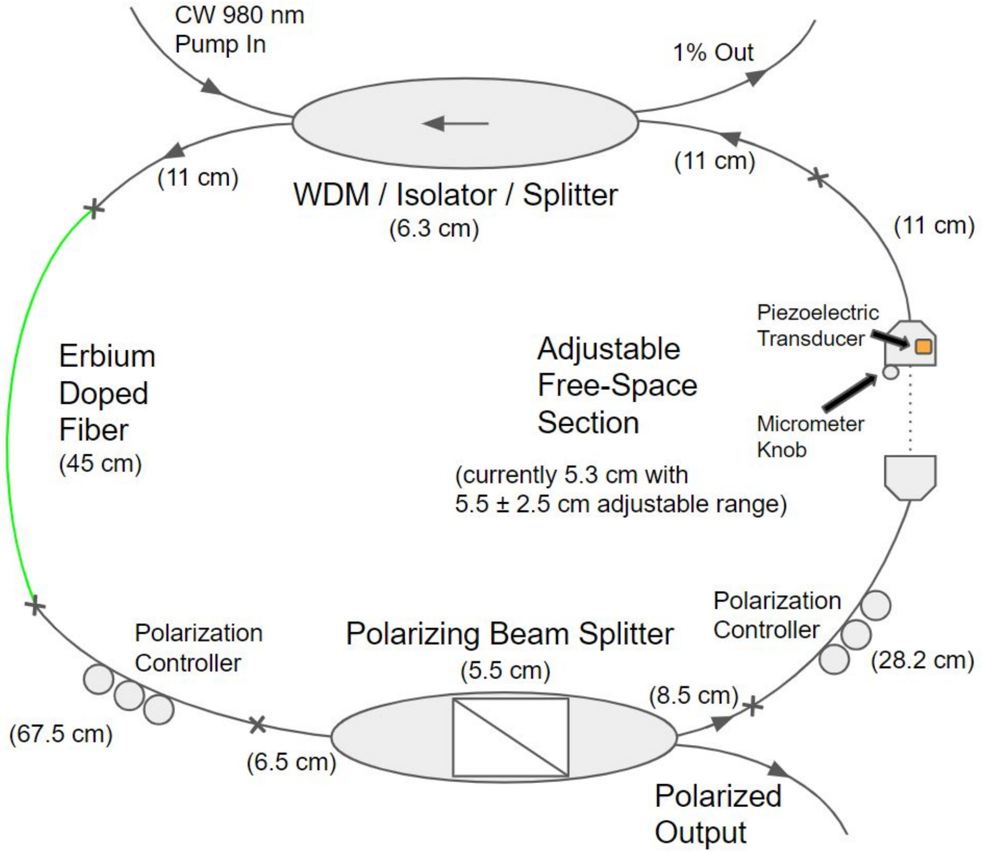

FIGURE 3.4: The adjustable free-space section features two FiberPorts which can be moved closer or farther for coarse length adjustments. A Micrometer knob and piezoelectric transducer is also present for fine adjustment of length. Several Components have been consolidated into a combination WDM/Isolator/Splitter and forward pumping is now used.

- **Vibrations**: Vibrations and mechanical disturbances in the laboratory can also affect the stability of the repetition rate by shaking the frequency comb causing sharp alterations in polarization or cavity length.

- **Noise from pump**: The pump laser used to pump laser light into the laser cavity can also introduce noise and power fluctuations within the cavity, which can affect the repetition rate.

- **Environmental changes**: Changes in the ambient temperature of the laboratory, including humidity or air pressure, will affect the repetition rate of the laser.

To ensure the stability of the repetition rate long term, it is important to carefully design the experimental setup and take appropriate measures to mitigate the effects of these factors. This can include using temperature stabilization, isolating the setup from mechanical vibrations (such as with an optical table), and controlling the ambient environment. Failure to properly stabilize the repetition rate can lead to reduced precision of measurements or loss of mode-lock.

3.2.1 Passive Stabilization

The optical frequency comb has been placed on an optical table in order to dampen any vibrations in the environment which may affect the comb. The frequency comb has also been placed in an aluminum box fastened to the optical table to further protection, ensuring that any air currents or sudden temperature changes in the environment will have negligible effects.

Inside of the aluminum enclosure, several thermoelectric tiles have been installed and soldered together to be controlled by an external temperature controller to aid in keeping the temperature stable. A thermistor has been installed inside of the enclosure in order to monitor the temperature based on the resistance so the controller can adjust accordingly. In order to more efficiently dissipate heat, a heat sink compound was also applied to the tiles, in addition to water cooling lines which have been installed beneath the enclosure.

3.2.2 Active Stabilization and Manual Tunability

In order to match the repetition rate of the adjustable repetition rate frequency comb with another frequency comb, it is required that repetition rates to be within approximately 1 kHz[8] (to avoid loss of spectral resolution), which requires the fiber lengths of the combs to be within approximately 50 μm. When cleaving fiber during preparation for splicing, lengths were able to be cleaved accurately within approximately 1 mm, so more precision is needed. The various methods of adjusting the length of the free-space section can provide the necessary precision. The primary method of making coarse adjustments to the length of the free-space section was by moving the FiberPorts along the guide rails. The 8 cm long guide rails provided approximately a 5 cm range of movement with a minimum length of 3 cm and a maximum length of 8 cm. Although this provides a good coarse adjustment of the length for precision of approximately 0.5mm, more precision can be obtained by using the micrometer knob. The micrometer can make very fine adjustments and can adjust the repetition rate on the sub-kHz level. The main drawback of using the micrometer is that it has a very small range of motion (approximately 1mm), but effective length adjustments of the FiberPorts can help alleviate this restriction. For the finest adjustments, a piezoelectric transducer has been installed inside the micrometer knob which can provide high-bandwidth tuning within a 100Hz range. The piezoelectric transducer can be controlled remotely while the comb is fully enclosed, and can be used with a phase-lock loop to lock the repetition rate to a frequency synthesizer or other reference. The final optical frequency comb design in conjunction with the stabilization methods can be seen in Figure (3.5).

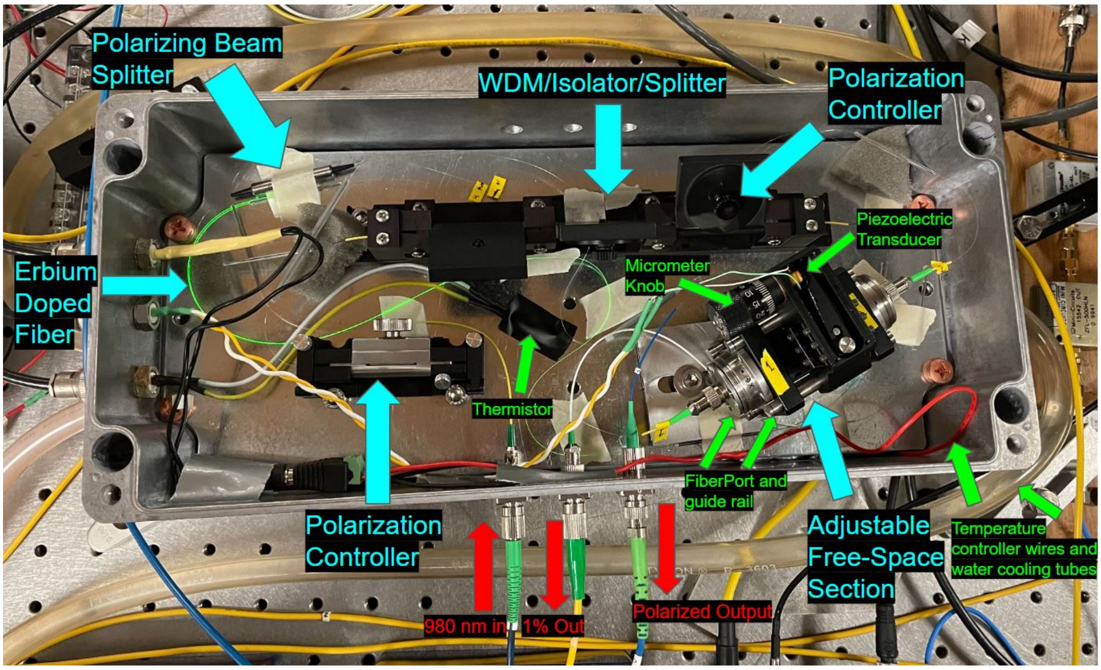

FIGURE 3.5: The optical frequency comb, mounted inside an aluminum enclosure with temperature control tiles and water cooling underneath.

3.3 Methods of Testing

After being constructed, the adjustable repetition rate frequency comb must be calibrated by performing some preliminary tests to ensure that the comb is functioning properly. The main method of testing the frequency comb was spectroscopy of a sample of $H^{13}C^{14}N$. Light from the frequency comb was sent through a container of HCN and analyzed by an optical spectrum analyzer according to the setup in figure (3.6). HCN is a linear molecule which has known absorption lines at certain wavelengths in the 1550 nm range[40], so it is good candidate for testing. The spectrum of the frequency comb was measured before the light passes through the sample as well as after passing through the sample and the ratio of the two spectra was analyzed for any differences. We would expect to see a dip in the spectrum at wavelengths of light that HCN absorbs which would be present in the light which passes through the HCN container that had not been present in the original spectrum.

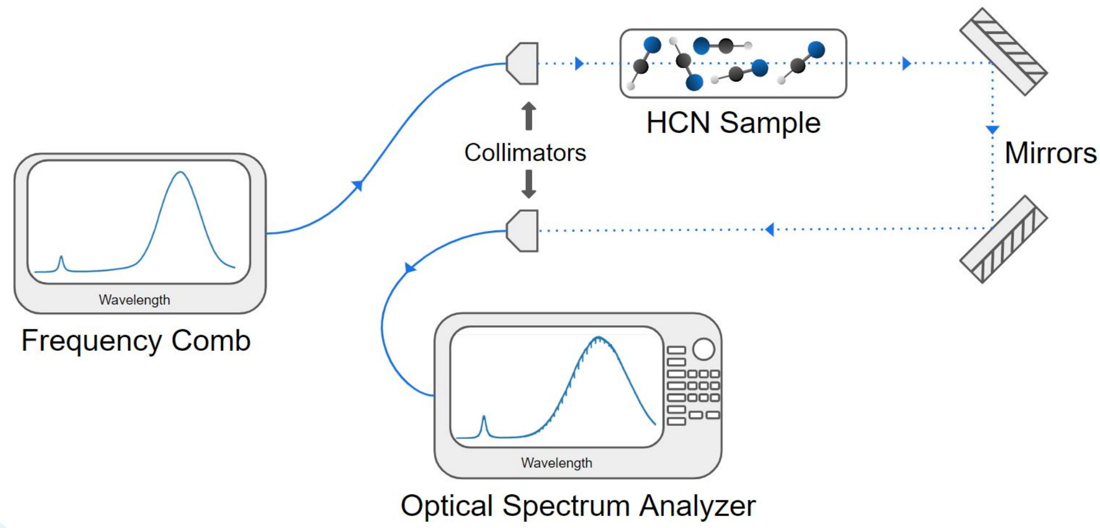

FIGURE 3.6: Light from the frequency comb was passed through a sample of HCN and reflected off a series of mirrors to be collected and analyzed in an optical spectrum analyzer.

Chapter 4

Results

4.1 Stabilization Data

The frequency comb was mode-locked and data was taken over several days of the room temperature as well as the repetition rate of the frequency comb as seen in Figure (4.1).

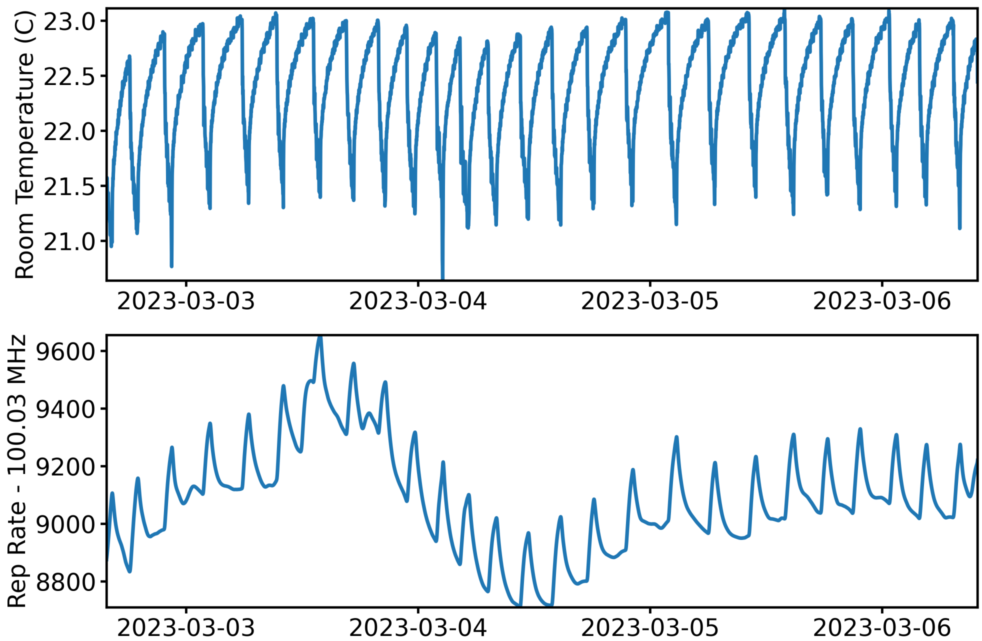

FIGURE 4.1: Graph of the temperature in the room (top) compared to the repetition rate of the frequency comb (bottom). The axes on the bottom left denote fluctuations from 100.039200 MHz ± 400 Hz.

The temperature in the room tended to fluctuate within 1° C of 22.0° C, while the repetition rate had short term drifts of 100-200 Hz and long term drifts of less than 1 kHz. Each sharp increase in temperature of the room corresponded to a sharp decrease in the repetition rate. For example, the first sharp increase in temperature from 21° C to 22.5°C caused the repetition rate to decrease by approximately 100 Hz, which is expected due to heat expanding the fibers and slightly increasing the time it takes a pulse to travel around the cavity. The temperature stabilization techniques which were implemented were able to successfully prevent the frequency comb from losing mode-lock over the long term.

4.2 Frequency Comb Spectrum and Preliminary Spectroscopy

The light from the optical frequency comb was sent into an optical spectrum analyzer in order to identify the spectrum that was produced. Light from the comb was then sent through a sample of $H^{13}C^{14}N$, a molecule with known absorption wavelengths[40], in order to determine if the frequency comb data would agree with the known absorption wavelengths. Graphs were compiled of the full optical frequency comb spectrum, zoomed in comb spectrum, zoomed in comb spectrum through $H^{13}C^{14}N$, and transmission data as seen in Figures (4.2), (4.3), (4.4), and (4.5) respectively.

The full width at half maximum of the frequency comb spectrum was measured to be 15 nm, although spectral features can be seen in a broader range of 1560±60 nm. There is a notable sharp peak at approximately 1530 nm in our spectrum, which is likely present due to the Er^{3+} ion favoring emission in the 1530 nm range[41]. Preliminary spectroscopy of the absorption wavelengths agree with the known absorption wavelengths within 0.05 nm. The significant distortion on the left side of the transmission graph, Figure (4.5), is likely due to the low signal of the frequency comb at wavelengths between 1540 nm and 1545 nm. Any noise that is present in the in both the comb spectrum as well as the comb spectrum through HCN will have significantly more effect on the ratio between the two spectrums at low power, resulting in very high noise in the 1545 nm region.

The most significant result is that the frequency comb is functional and can perform spectroscopy accurately. The laser remained mode-locked over a long period of time, which is necessary to be able to perform experiments in the future. The overall optical frequency comb spectrum is “clean” in the sense that there is no continuous-wave (CW) breakthrough present in the spectrum. CW breakthrough occurs when a continuous-wave laser signal, such as the one pumping light into the frequency comb, leaks into the comb spectrum. CW breakthrough can significantly degrade the quality of the comb spectrum, as it can mask the comb lines and introduce noise and distortion. The

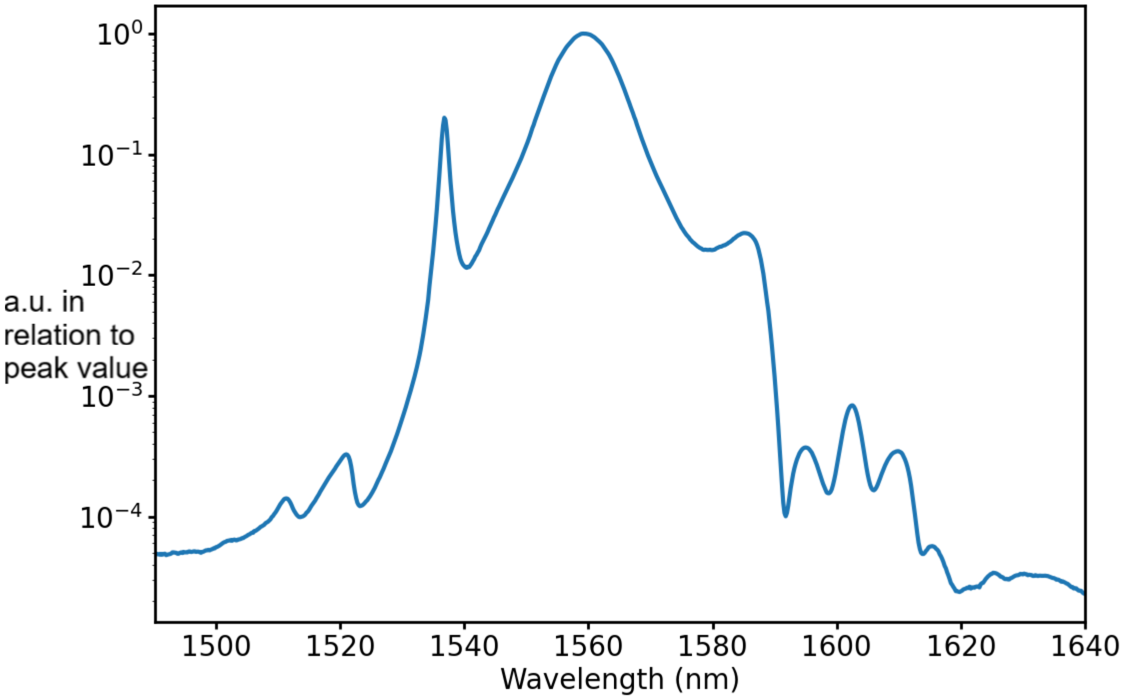

FIGURE 4.2: The full spectrum of the optical frequency comb on a logarithmic scale. FWHM = 15 nm.

frequency comb was also able to accurately scan a wide range of wavelengths and accurately measure absorption features of HCN in a 25 nm range, indicating good potential for spectroscopy in the future.

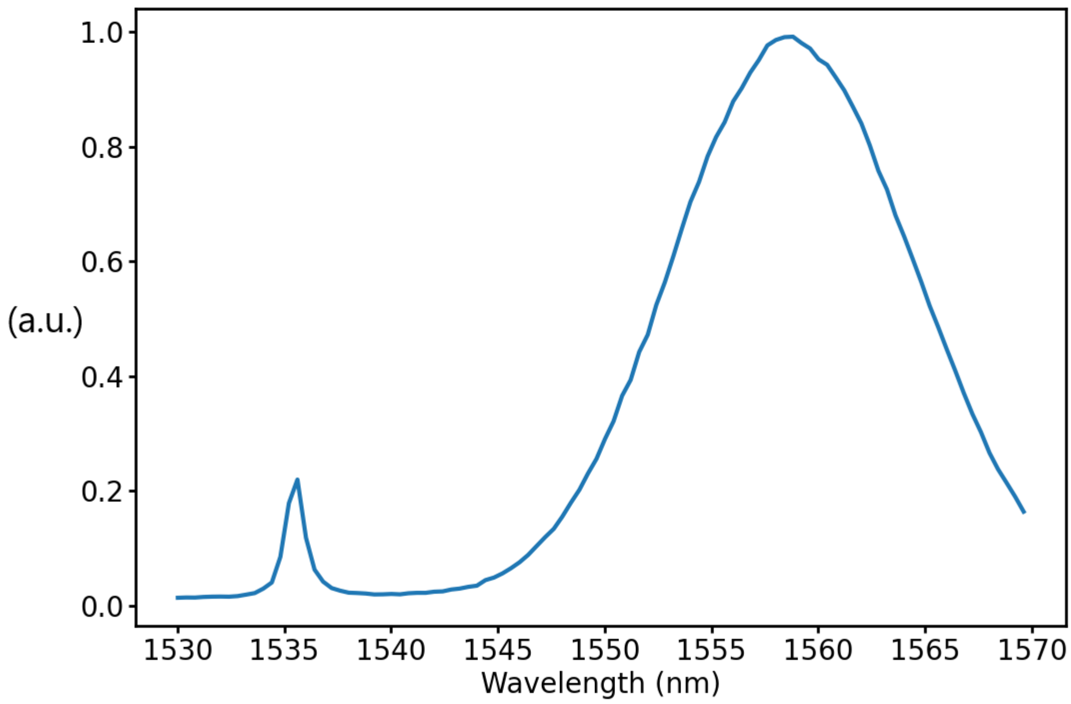

FIGURE 4.3: The zoomed in spectrum of the optical frequency comb on a linear scale.

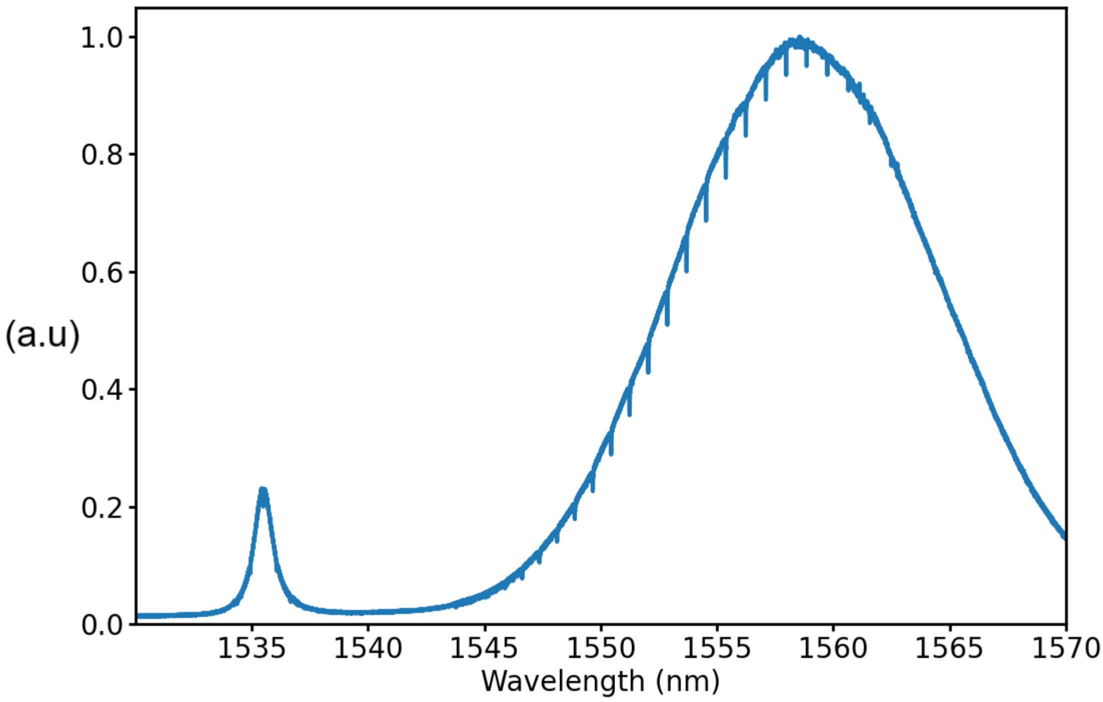

FIGURE 4.4: The zoomed in spectrum of the optical frequency comb through a sample of $H^{13}C^{14}N$.

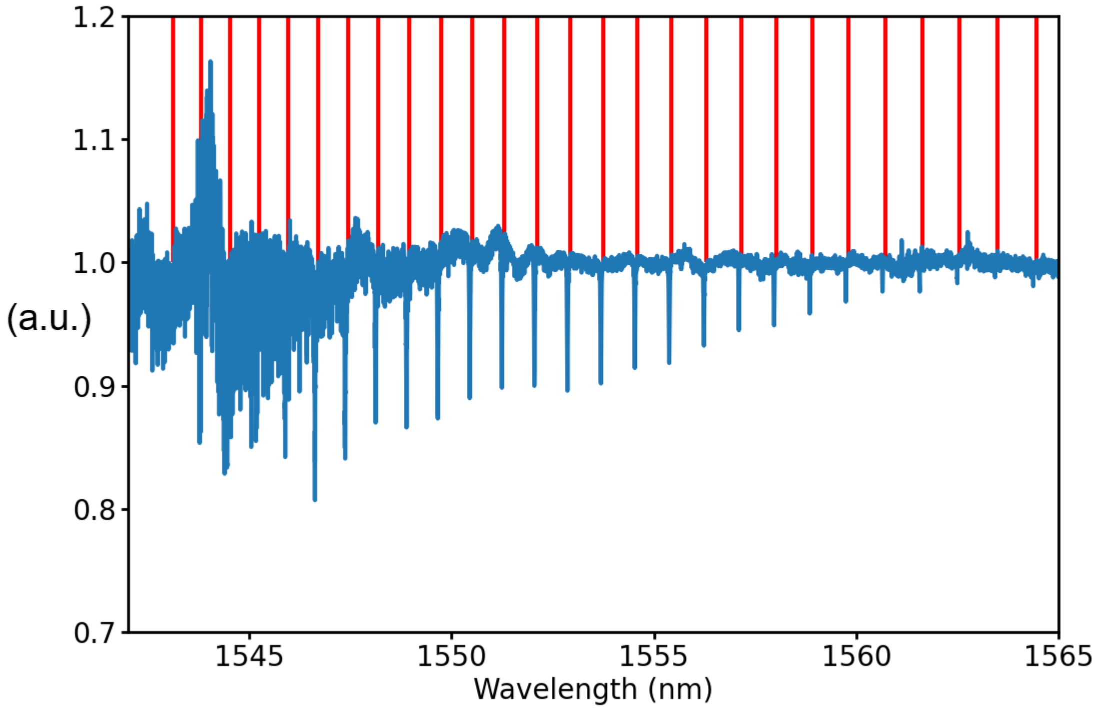

FIGURE 4.5: The transmission (blue) represents the ratio of the comb spectrum to the comb spectrum through $H^{13}C^{14}N$. Sharp dips in the spectrum represent absorption of light by HCN, as compared to known linecenters[40] (red).

Chapter 5

Future Directions

5.1 Dual-Comb Spectroscopy of Benzene

In order to perform dual-comb spectroscopy of benzene, several steps must be taken after the completion of the adjustable repetition rate frequency comb. After ensuring that the adjustable repetition rate frequency comb is functional, its repetition rate ideally must be matched within at least 1 kHz of the second optical frequency comb. The signal from the two combs must be interfered to ensure that the proper dual-comb signal can be seen. Once a dual-comb signal has been achieved, the spectrum must be broadened to the range at which benzene absorbs, which is in the 1650 nm range[10], using highly nonlinear fiber. In addition, the frequency comb signals must be stabilized to a narrow linewidth CW laser so that there is a reference for the comb teeth. Following these steps, dual-comb spectroscopy can be performed via a supersonic beam apparatus.

5.1.1 Dual Comb Signal

Light from the adjustable repetition rate frequency comb was mixed with the light from a previously built frequency comb with both combs having a repetition rate of 100.038 MHz and a difference of repetition rate of approximately 100 Hz. The intefereogram produced by the dual-comb signal can be seen in Figure (5.1).

Although a dual-comb signal was produced, it did not particularly provide very much information, and no spectroscopic information was able to be retrieved. The interfereogram shows that the spectra of each optical frequency comb do overlap, meaning that comb teeth from one comb will interfere with comb teeth from the other comb. Although not a completely successful preliminary dual-comb signal, it is the first step on the road to a better measurement, and steps can be taken to improve the signal in the future.

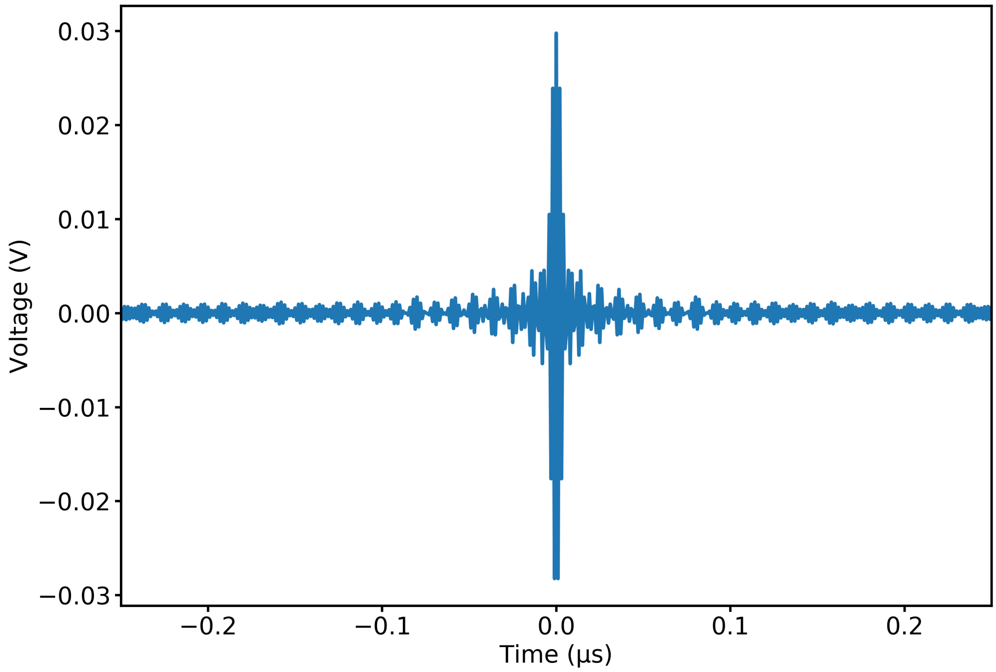

FIGURE 5.1: Dual-comb interfereogram with repetition rate filtered out.

Comparing to an interfereogram from a successful experiment performed by Schliesser et al.[42], there is a cleaner interferogram with spectral information of NH_3 contained in the dual-comb signal.

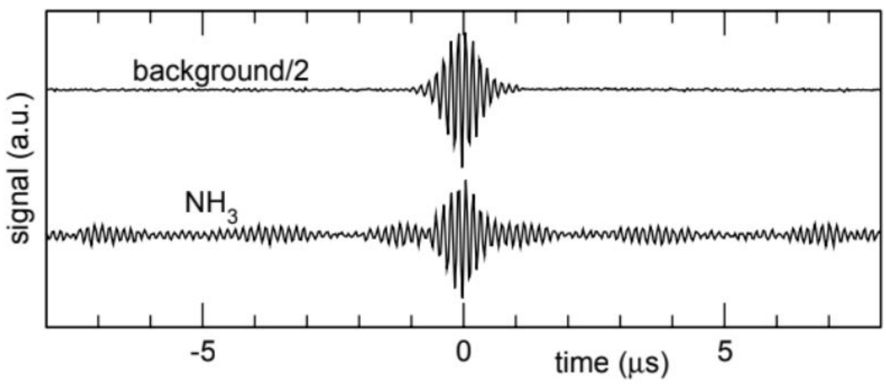

FIGURE 5.2: A dual-comb signal (top), compared withthe interfereogram from dual-comb spectroscopy of NH_3 (bottom) from Schliesser et al. (2005)[42].

5.1.2 Spectral Broadening

The spectrum of the optical frequency comb is centered at 1550 nm, and has a full width at half maximum of 15nm. Such a spectrum is not at the right range of frequencies to perform spectroscopy of benzene, which has known absorption wavelengths at approximately 1650 nm[10], so spectral broadening must be achieved to move the spectrum to cover a range around 1650 nm. Several spectral broadening techniques can be used to move the spectrum, one of which involves splicing to highly nonlinear fiber (HNLF) and the other which involves an erbium doped fiber amplifier (EDFA).

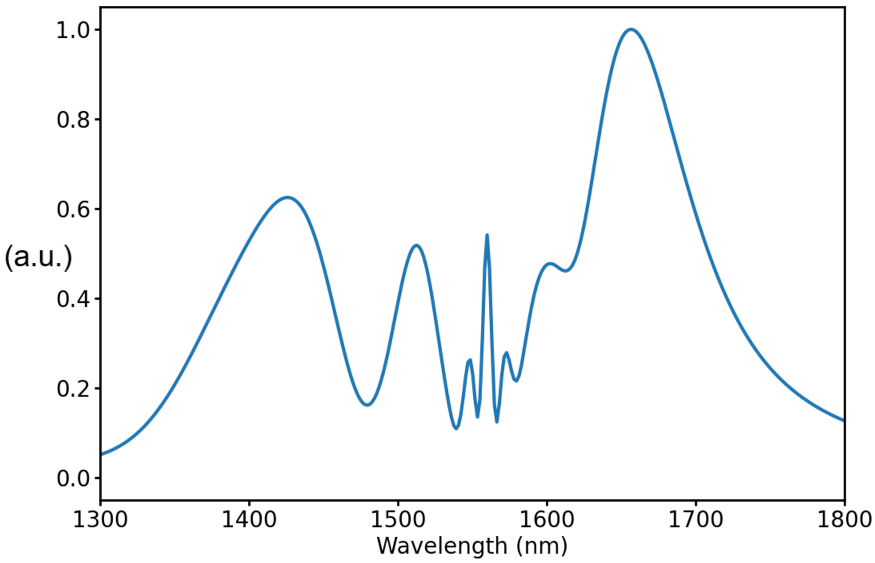

FIGURE 5.3: Simulation data using a section of highly nonlinear fiber (30 cm), a spectrum of light centered around 1550 nm, with a 200 fs pulse and 80 mW average power, will be broadened and encompass a range of wavelengths around 1650 nm, precisely where benzene has known absorption wavelengths[10].

Highly nonlinear fiber (HNLF) can be used to broaden the spectrum of an optical frequency comb through a process known as supercontinuum generation[43]. This process involves the nonlinear interaction between a broad spectrum of light and the fiber's material properties, resulting in the generation of new frequencies and a broadening of the initial spectrum. In addition, the dispersion properties of the HNLF also play a role in the broadening of the comb spectrum. The dispersion properties of the fiber cause the different frequencies within the comb to travel at different velocities, resulting in a temporal broadening of the pulses. This temporal broadening leads to an increased overlap between the different frequency components, which in turn enhances the nonlinear interactions and further broadens the spectrum[44].

Another method of spectral broadening is through the use of an erbium doped fiber amplifier[45]. EDFAs are a type of optical amplifier which is used to amplify the signals in optical fibers, such as from an erbium fiber laser. EDFAs are useful in conjunction with optical frequency combs as the power of each comb tooth may be too low for accurate spectroscopy to be performed[46]. Amplification of the frequency comb teeth will help reduce the relative noise of the signal as well which is necessary for accurate measurements. In addition, EDFAs provide a wide amplification bandwidth, which is necessary if there is low power at wavelengths which benzene absorbs.

5.1.3 Optical Stabilization

In order to reduce noise and increase precision of spectroscopic measurements, the comb teeth of the optical frequency must be stabilized[3]. The teeth of the optical frequency comb need to be referenced to some external frequency, or the teeth will drift over time leading to reduced resolution[47]. Returning to the frequency comb equation (2.1),

$$f_n = nf_{rep} + f_0,$$

despite efforts to stabilize the repetition rate, the factor of n, which is generally on the order of 2×10^6, will amplify any instability in the repetition rate when looking at the radio frequency during dual-comb spectroscopy. In addition, the offset frequency f_0 needs to be accounted for. By using a CW laser with a narrow linewidth (ideally on the order of 1-10 KHz), a reference can be provided for the comb teeth. The wavelength of the laser must be in the 1550 nm range to ensure that it is between two comb teeth so that the spacing between the reference laser and the two adjacent comb teeth can be measured. The narrower the linewidth of the CW laser, the more well-defined the spacing between the two adjacent comb teeth will be, providing much higher stability and precision as well as accounting for the offset frequency.

In order to reduce the linewidth of a CW laser as much as possible, it is necessary to build an external cavity diode laser (ECDL). The external cavity provides an additional level of feedback that can be used to stabilize the laser frequency[48]. The length of the external cavity in conjunction with the use of a grating is adjusted to create an interference condition that selects a single frequency of the laser diode in the external cavity. The grating acts as a dispersive element, separating the various frequencies emitted by the laser diode. By selecting a specific order of diffraction, the laser can be tuned to a precise wavelength, reducing the linewidth even further[49].

5.1.4 Supersonic Beam Apparatus

A supersonic beam apparatus is often used in spectroscopy using optical frequency combs because it provides a controlled environment to study the molecules of interest[50]. Using this technique, the benzene molecules will be mixed with argon to create a gaseous sample of benzene, which can be pumped into a vacuum chamber vertically downward. The light from the dual optical frequency combs can enter horizontally into the chamber, perpendicular to the benzene molecules. Using a supersonic beam apparatus provides an advantage over using a simple container of a sample, such as was used with HCN, because the supersonic beam apparatus is able to cool molecules into their lowest energy state and isolate individual molecules to ensure that their properties are not affected by collisions with other particles or thermal motion[51], resulting in a cleaner spectrum with sharper lines, making data easier to analyze and interpret. Another advantage is that the isolation of molecules in the supersonic beam apparatus allows for the study of individual molecules of benzene, rather than a bulk sample, which can be important for understanding molecular structure and behavior of the molecule in a certain environment. Currently, the supersonic beam apparatus in the lab has successfully been able to reduce the pressure inside the vacuum chamber to approximately 10^{-5} Torr, although it will not be clear if less pressure will be needed until testing is performed to determine whether the benzene will be in a low enough energy state.

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Appendix A

Fiber Components

Components used in the final design of optical frequency comb (Figure (3.4)). All fiber splices between components were performed using an Ericsson FSU 995 PM Fiber Fusion Splicer.

- **WDM/Isolator/Splitter**: Oplink WTIH WDM / tap coupler / isolator hybrid combination (WTIH1598S012111G).
- **Polarizing Beam Splitter**: Comcore 2x1 Fused PM Fiber Standard Combiner.
- **EDF**: LIEKKI®Er110-4/125 fiber.
- **Polarization Controllers**: Thorlabs FPC030 - Fiber Polarization Controller, 3 Ø27 mm Paddles and Thorlabs CPC250 - In-Line Fiber Optic Polarization Controller for Ø250 µm Bare Fiber.
- **Collimators**: Thorlabs PAF2-2A - FiberPort, FC/PC & FC/APC, f=2.0 mm, 350 - 700 nm, Ø0.33 mm Waist. Used in conjunction with Cageplate adapters.
- **Free-Space Adjustment**: Thorlabs SM1ZA - Z-Axis Translation Mount, 30 mm Cage Compatible. Thorlabs PC4FL - Co-Fired Piezo Actuator, 4.6 µm Max Displacement, 5.0 mm x 5.0 mm x 5.0 mm.

HCN cell used:

- Wavelength References HCN-13-T(25x5)-25-1550 Cell filled with 25T pure C13 HCN. Cell has 25mm OD, 5cm path length with B270 glass optics AR coated for 1550nm.