Dual Beam Frequency Comb FTIR Spectroscopy

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DUAL BEAM FREQUENCY COMB FTIR SPECTROSCOPY

by

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ABSTRACT

A visible and Infrared (IR) range dual beam frequency comb Fourier transform spectrometer was developed. Using dual mode-locked Ti:Sapphire lasers a comb-interferogram was generated in the visible range. This spectrum was calculated and used to measure the transmittance of a Nd-doped crystal. The system was further developed to generate an IR interferogram by Difference Frequency Generation (DFG) using a Gallium Selenide (GaSe) crystal placed in the mode-locked pump beam. Numerical work was done to calculate the expected DFG spectrum confirming the necessary IR range can be reached. This has been conducted in support of an IR holographic spectroscopic microscopy spatial and spectral resolving platform for applications in cell biology and biomedical applications.
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INTRODUCTION

This work has been done in support of an applied Comb-FTIR and holographic pathogen identification platform to be used in conjunction with an artificial intelligence which will provide data for devices using spectral data for onsite pathogen identification. The major steps in the process is the use of a 2D holography microscope to identify malignant versus non-malignant cells. This spatial data is then coupled to the spectral data from the comb-FTIR and sent to the AI. The AI then analyzes the spectral information and finds the defining characteristics of that spectral signal to be sent to portable and or cheaper devices for the identification of cells onsite at testing facilities.

This thesis is primarily focused on the creation of a stable comb-FTIR for the generation of the spectral information. Figure 1 provides a schematic of the optical layout. The Ti:Sapphire laser and pump is in a safety interlock controlled apertures for safety. The pump beam is controlled through a pair of alignment mirrors controlled via piezos. The cavity is also externally controlled through a radio frequency tone generator with a set of slaved proportional integral derivative (PID) controllers that directly alter the cavity length through stepper and piezos. Following the emitted beam, we see after passing through a beam splitter that 5% of the beam power is directed to a calibrated spectrometer and Indium Gallium Arsenide (InGaAs) detector after passing through a beam combiner. The remaining 95% of the beam is directed through a
GaSe based difference frequency generation and eventual Zinc Selenide (ZnSe) beam combiner and a liquid nitrogen cooled infrared sensitive Mercury Cadmium Telluride (MCT) detector.

Figure 1 Optical Layout

The Ti:Sapphire oscillators pumped by a 10 W green laser. The pump and oscillator beam path are shown in Figure 2.
Figure 2 532 nm Pump and Ti:Sapphire Oscillator

p labels are pump beam mirrors and m are mode-locking mirrors.

The green pump is beam split and provides roughly ~4.5 W of power for each oscillator channel. Once tuned, the cavity allows mode-locking to be achieved to produce a steady train of nominally 10 fs pulses of roughly ~450 mW average power as measured by a Si photodetector.

A two channel, low phase noise tone generator, acting as a master oscillator provides the 8th harmonic of a 124MHz tone to allow PID control of the oscillator frequency to maintain a steady difference, or “beat note” frequency. Using a high-speed beam combiner, the two beams are made co-linear and focused on a 100 MHz InGaAs detector to generate an interferogram at the beat note frequency. The Fourier transform of this interferogram leads to the spectral content of the mode-locked laser pulses, spanning from roughly 650nm to 950nm.¹ This spectroscopic system is characterized by comparing the known absorption lines of a Neodymium doped
Strontium Fluorovanadate (Nd:SVAP) laser crystal, which is then compared to published FTIR measurements.  

Further work is to develop the necessary knowledge base and calculations to implement a GaSe based Difference Frequency Generation (DFG) apparatus. Phase matching the red incident electromagnetic waves to the generated infrared in the non-linear uniaxial crystal. The method employed for this calculation relies on assumptions of the experimental apparatus limitations and the useful ranges of values desired for implementation in the work that this experiment supports. From these calculations we optimized the response to obtain the desired 8 μm – 14 μm spectrum.  

The goal is to provide infrared interferograms as part of a holographic spectroscopic microscopy spatial and spectral resolving platform. The application of this device will be for the identification of cellular pathogens specifically cancerous cells.
Lasers have a boundary condition defined by the presences of zero amplitude (nodes) at the end mirrors of the cavity. To illustrate this Figure 3 presents a schematic of a laser cavity with several of the lowest order standing waves that satisfy the boundary condition. Clearly, an infinite number of these “longitudinal” modes are possible. When there is a gain medium in the cavity, only those laser modes that fall under the gain curve of the crystal will lase. Every laser operates only in the spectral range where there is gain, and this determines the laser emission spectrum. Because of mode competition, a laser generally has a narrow, continuous wave (CW), spectrum, with a very narrow spectral line width. We can expect with the use of Ti:Sapphire for the intersection of the gain curve and modes to have a 800 nm centered wavelength when continuous wave lasing occurs. Mode locking occurs when all allowable laser modes oscillate in phase. When mode locking is achieved then the emission spectrum is broadened, potentially up to the limits of the gain spectrum. For Ti:Sapphire with its broad range of modes and gain curve we expect to see a spectrum from 700 nm to 900 nm ideally.
For lasers in general the wavelengths of the allowed longitudinal cavity modes are \( \lambda_n = \frac{2L}{n} \) where \( L \) is the length of the cavity and \( n \) the integer that gives the number of half-wavelengths in the cavity for that mode. If the cavity includes optically dense media, then \( L \) stands for the optical length of the cavity, which is the physical length multiplied by the refractive index of the medium between the mirrors. The frequency of oscillation \( f_n \) for the electromagnetic fields of the allowed modes is related to their wavelength by.

\[
f_n = \frac{c}{\lambda_n} = \frac{c}{\frac{2L}{n}} \quad (1)
\]

\[
f_n = nf_r \quad (2)
\]

The roundtrip frequency for a photon in the cavity is given by \( f_r \). Every photon in every mode has this round-trip frequency and a round trip time of \( \tau_r = \frac{2L}{c} = \frac{1}{f_r} \). However, the field in each mode oscillates at different and much higher frequencies \( f_n \) given by Equation 1. The total field in the cavity is the linear superposition of all modes present, which gives a complicated but periodic field with a large number of oscillations at all possible differences between the various
mode frequencies. The lowest possible difference is the difference in the frequency of neighboring modes where n differs by unity, which is the cavity round trip frequency \( f_r \).

For mode-locking the total laser field is the superposition of all allowed modes with different amplitudes and phases, expressed in the following equation

\[
E(t) = \sum_{n=N}^{M} A_n \cos(2\pi f_r t + \phi_n). \tag{3}
\]

To illustrate the effect of mode-locking, we set \( f_r = 10 \) Hz, and take different sums from \( N = 10 \) to \( M = 10 + m \), with \( m = 0, 1, 2, 3, \ldots 9 \). These numbers are set artificially small to simplify the calculation and to allow for clear and illustrative figures. The lowest oscillation frequency would be \( 10 \times 10 \) Hz = 100 Hz. The highest would be 190 Hz. For the actual laser cavity, we will be using, the round trip frequency will be \( \sim 124 \) MHz, and the field oscillations will be as high as \( \sim 375 \) THz, so that the values of \( N \) and \( M \) are of order 3 million.

In Figure 4, we plot intensity vs time for a time range of 0 to 2.5 round trip periods \( \tau_r \), for a total time of 250 ms. Intensity is proportional to the square of the field amplitude, which is the actual quantity plotted in Figure 4, according to

\[
E(t)^2 = \left[ \sum_0^m \cos(2\pi(10+m)10t) \right]^2. \tag{4}
\]

The effect of mode-locking is to make all \( A_n \) and all \( \phi_n \) equal. We take \( A_n = 1 \) and \( \phi_n = 0 \) for all modes. We first choose \( m = 0 \). This corresponds in Equation 4 to a single mode oscillating at 10 Hz, as shown in Figure 4a for Figure 4b, we let \( m = 1 \), which gives us a superposition of two modes, at 10 and 11 Hz. The interference frequency of \( f_r = 10 \) Hz appears as a slow modulation of the amplitude of the fast “carrier”. With \( m = 2 \), we have the superposition and interference of 3 modes, shown in Figure 4c. The last is the superposition of 10 modes, shown in Figure 4d, where the laser output already has the appearance of a stream of narrow pulses with period \( f_r \).
Importantly for non-linear optics applications, the peak intensity of the mode-locked superposition of 10 modes is $100 \times$ higher than the intensity of a single mode.

![Figure 4 Numerical Mode-Locked Laser Simulation](image)

The temporal duration of the laser pulse $\Delta t$ and width of the emission spectrum $\Delta f$ is limited by the relation

$$\Delta t \Delta f \sim 1 \quad (5)$$

Thus, a laser that oscillates continuously, so that $\Delta t$ is very large, can have a very narrow spectral width $\Delta f$, which might comprise just a single longitudinal mode. On the other hand, a short duration laser pulse with small $\Delta t$, must have a large $\Delta f$. This situation is indicated schematically in Figure 5. The CW laser has only a few cavity modes. The short pulsed laser has a spectrum of many more modes. This was observed already in simulation of Figure 4, where inclusion of more modes in the sums, i.e. with broader frequency range, gave temporally narrower pulses. Each
short pulse output by a mode-locked laser individually comprises a broad spectrum, and each pulse has the same broad spectrum. Let’s suppose for Figure 5 that the mode separation is 124 MHz, and that the laser pulse is “long”, e.g. 1 ns. Then according to Equation 5, the laser emission spectral width would be 1 GHz, which exceeds the $f_r$ mode separation by 8×, so that the emission spectrum comprises 8 modes. If we reduce the pulse width to 100 ps, the emission spectral width and number of modes both increase to 10 GHz and 80 modes, respectively. Next, suppose a mode-locked laser with 20 fs pulses, giving a laser emission spectral width of 50 THz. Now, the number of oscillating modes has increased to 400000, which is impossible to show graphically. Figure 5 therefore represents a harmonic frequency comb with evenly space modes each an integer multiple $f_r$. 
Because the mode-locked red lasers comprise a frequency comb a frequency offset at zero frequency defined as “carrier offset frequency” or “carrier envelop offset”, given by the symbol $f_o$. $f_o$ is a result of the dispersive medium of the passive Ti:Sapphire crystal having a frequency dependent refractive index. This is the difference between an evacuated cavity and a non-evacuated cavity. The evacuated cavity will have no dispersive medium and the frequency vs mode number will be a linear relationship that extrapolates down to (0,0). The non-evacuated cavity in this case a passive Ti:Sapphire crystal is instead now a non-linear function of the mode number relationship and extrapolates to a number not equal to (0,0). Continuous wave lasing of the Ti:Sapphire will still have an offset that may differ from the passive case. Mode locking

Figure 5 Harmonic Frequency Comb
(a) 1 ns and (b) 100 ps pulses
however forces the frequency vs mode number function to be linear. This new linear relationship
will still not extrapolate back to (0,0) due to a new difference between modes as compared to the
evacuated case and instead to a number \( f_0 \). \( f_0 \) while present in the single modelocking case
becomes far more concerning when the interferogram of two superimposed beams is measured
and will be elaborated on in Chapter 2.

The experimental results regarding lasing both CW and mode-locking have been
observed from the actual Ti:Sapphire lasers being used to perform this experiment. Figure 6a
shows a spectrum measured directly by a spectrum analyzer of the beam for CW (dotted line)
and mode-locked (solid line). We can easily observe the narrow versus broad emission spectrums
for CW and mode-locked lasing, respectively. Figure 6b is the measurement from an
autocorrelator of the resulting pulse duration of 9.3 fs.

![Figure 6 CW and Mode-locked Lasing Spectrum and Pulse Duration](image)

For lasers in the infrared and visible spectral range, detectors are usually too slow to
directly observe the fast oscillations of the electric field at multiples of \( f_0 \) in the laser output. As
all signals measured within this experiment are in those ranges we can only rely on measuring
and discussing signals measured through methods that do not use direct measurements of the electric field and so we will rely on the generation of interferograms by interference between mode-locked pulse trains and the scaling of those interferograms to recover optical information.
CHAPTER TWO: DUAL BEAM FREQUENCY COMB SPECTROSCOPY

We next consider interferograms produced by the superposition of two mode-locked laser beams. This second laser will be assumed to function identically to the first, except that its cavity length is slightly longer, giving rise to a round-trip frequency \( f'_r \) that is slightly different by an amount \( \Delta \).

\[
f'_r = f_r + \Delta \tag{6}
\]

This is achieved, for \( \Delta > 0 \), by having a cavity length for the second laser that is smaller than that for the first laser, namely \( L' < L \). Then,

\[
\frac{1}{f'_r} = \frac{1}{f_r + \Delta} = \frac{2L'}{c}, \tag{7}
\]

The frequency \( \Delta \) offset will be small compared to the roundtrip frequency \( \Delta \ll f_r \). The modes of each laser are

\[
nf_r \tag{8}
\]

\[
mf'_r = m(f_r + \Delta) \tag{9}
\]

for the first and second laser, respectively, where \( n \) and \( m \) are integers. When the two lasers are overlapped in space, and when the mode-locked pulses overlap in time, every mode of one laser will interfere with every mode of the other laser. For example, modes \( nf_r \) and \( nf'_r \) will interfere to produce an interference frequency \( n\Delta \). Similarly, \( (n + 1)f_r \) and \( (n + 1)f'_r \) will produce an interference frequency \( (n + 1)\Delta \), and so on. There will also be interference between modes \( nf_r \) and \( (n + 1)f'_r \), producing interference frequencies \( f_r + (n + 1)\Delta > f_r \), but such interferences would be at much higher frequencies than those produced by interference of modes with \( m = n \). We will show that those interference frequencies cannot be “sampled” by a stream of mode-locked pulses at \( f_r \), and to avoid “aliasing” artifacts those fast interferences need to be
electronically filtered out. The same applies to interference of any modes with larger differences between m and n. A key point is that, because \( \Delta << f_r \), the interference frequencies with \( m = n \) namely \( n\Delta, (n + 1)\Delta, (n + 2)\Delta \) are all much lower than any mode frequencies given by Equations (8,9), even though the values of n for our Ti:Sapphire laser are on the order of 3 million.

Figure 7a illustrates the appearance of interference frequencies in integer multiples of \( \Delta \). We assume an overly simplified model, in which \((n,m)\) take the values 1-5. Modes from the two lasers with the same mode number interfere to produce interference frequencies of \( \Delta, 2\Delta, 3\Delta, 4\Delta, \) and \( 5\Delta \). While the mode-locked pulses of the two lasers temporally overlap, the amplitude of those pulses will be modulated at those interference frequencies. A Fourier transform of a transient recording of the superposed lasers then would reveal those interference frequencies as shown in Figure 7b.

Figure 7 Interference Frequencies
To demonstrate the modulation of the mode-locked pulse amplitudes, we present a simple numerical example based on Figure 7. We consider the time dependent electric fields of the two mode-locked lasers. All modes in each laser have the same phase \( \phi_n \) and \( \phi'_n \) due to mode-locking. We take this phase to be zero. Similarly, amplitudes of all modes in each laser \( A_n \) and \( A'_n \) are equal, and we take their values to be unity. Then

\[
E(t) = \sum_{n=1}^{5} \cos(2\pi n f_r t)
\]

(10)

\[
E'(t) = \sum_{m=1}^{5} \cos(2\pi m f'_r t)
\]

(11)

Each of these expressions represents a pulse train as in Figure 4, but with pulse repetition rates that differ by \( \Delta \), so that pulses drift in and out of phase with a period \( 1/\Delta \). When the pulses of the spatially overlapped beams coincide temporally, they will interfere. The time dependent intensity then will be

\[
U(t) \propto (E(t) + E'(t))^2 = E^2 + E'^2 + 2EE'
\]

The first two terms comprise the simple sum of two mode-locked pulse trains, where within each pulse the fields oscillate at “optical” frequencies that are too fast to be recorded or observed. The cross term \( EE' \) is the interference term, which includes terms such as \( \cos(2\pi n f_r t)\cos(2\pi n f'_r t) \). These give rise to terms at sum and difference frequencies, e.g. \( \cos[2n (f_r-f'_r)t] + \cos[2\pi n (f_r+f'_r)t] \). The difference frequency terms give the temporal modulations of the pulse intensities that are sufficiently slow to be observed. That modulation is given by

\[
U(t) = \sum_{n=1}^{5} \cos(2\pi (n\Delta)t)
\]

(12)

In other words, the intensity of the superposed temporally and spatially coincident mode-locked laser pulses will vary periodically in time with frequencies \( \Delta, 2\Delta, 3\Delta, 4\Delta, \) and \( 5\Delta \). Those variations are all much slower than the repetition rate \( f_r \approx f_r' \) of the superposed pulses and much
slower than the optical frequencies of individual longitudinal laser modes. The maximum factor by which they are slower than \( f_r \) is \( S = \frac{f_r}{\Delta} \), which is called the frequency down conversion factor.

Figure 8 shows pulses calculated from Equation 10 and 11 emitted by two lasers with \( f_r = 1 \text{ Hz} \) and \( f_{r'} = 1 \text{ Hz} + \Delta \), where \( \Delta = 0.1 \text{ Hz} \). The two pulse trains look identical except for the 0.0909 sec difference in their pulse repetition periods \( \tau \) and \( \tau' \). The first pulses at \( t = 0 \) overlap perfectly in time and would interfere when overlapped also in space. But the overlap is reduced for the second pulses, and consequently the interference would also be reduced. After the second pulse there is no longer any interference possible. After about 10 pulses, or a duration of \( 1/\Delta \), the pulses drift back into phase again and would interfere when spatially overlapped. Thus, interferograms appear with a temporal period of \( \Delta \), the “beat” frequency. An oscilloscope can trigger on the interferograms, so that its display can present each interferogram in sequence or the temporal average of multiple sequentially digitized interferograms. Similarly, a spectrum analyzer can trigger on the interferograms to produce the interference frequency spectrum within each interferogram or averaged over interferograms.
Figure 8 Simulated Time Overlapping Pulse Trains

In practice, $\Delta$ will be much smaller than 10% of $f_r$. Then a much larger number of pulses for the two lasers will overlap temporarily before the contributions from the two lasers drift out of synchronicity. The interference between them will be manifested as a periodic modulation of the superposed pulse amplitude, with periods given by $1/n\Delta$ according to Equation 12. The fastest observable modulation is presented in Figure 9(b), where each vertical line represents one of the temporally and spatially coincident pulses from the two lasers. The pulse heights are modulated at a frequency $f_r\Delta/2$. Any faster modulation would cause aliasing. Observing Figure 9(b) it is apparent that if the frequency were any higher aliasing would occur as the sampling theorem would then lead to a false frequency.

If $N_{max}$ is the highest mode number in the laser mode spectrum, then the fastest modulation would be $N_{max}\Delta$. To observe this modulation without aliasing imposes a limit on the
maximum useful value of $\Delta$, namely $\Delta = f_r/2N_{\text{max}}$. As a numerical example relevant to our experiment. We assume for now mixing of the two lasers beams produced by the Ti:Sapphire lasers in the red-to-near-IR spectral range. The shortest wavelength that can be amplified by the Ti:Sapphire crystal is 680 nm, which corresponds to an optical frequency of $\nu_{\text{max}} = 441$ THz. Then $N_{\text{max}} = \nu_{\text{max}}/f_r = 3.56$ million. Thus, with $f_r = 124$ MHz, the maximum useful value of $\Delta$ would be 17 Hz.

The minimum observable modulation frequency must also be considered. The lowest modulation frequency is determined by the condition that its period must be shorter than the interval of time where the pulses overlap sufficiently to interfere. To calculate the duration of pulse overlap we first divide the pulse duration by the difference in pulse period $1/f_r – 1/f_r'$ to obtain the number of pulses that overlap for every interferogram. Multiplying that by the pulse period gives the duration of overlap. The inverse of the duration of overlap gives the slowest observable modulation frequency (though this minimum may still be unobservable for another reason discussed below).

As an example, we take conditions relevant to our experiment. Suppose $f_r = 124$ MHz and $f_r' = 124$ MHz + 17 Hz. Suppose the pulse duration is 15 fs. Then the number of pulses that overlap would be $15 \times 10^{-15}/(1/124000000 – 1/124000017) = 15$. The duration of the overlap is then $15/124$ MHz = 120 ns. The minimum observable modulation frequency based on the criterion of temporal pulse overlap is therefore 8.3 MHz.

In the case of Ti:Sapphire lasers, a minimum modulation frequency of 8.3 MHz will not be observed. The minimum modenumber for the minimum modulation frequency is $8.2\text{MHz}/17\text{Hz} = 490000$, but this is much smaller than the minimum number of longitudinal
modes at the longest wavelength (890 nm) amplified by Ti:Sapphire. That number $N_{\text{min}}$ is 2.7 million. Thus, the smallest observable modulation frequency would be $N_{\text{min}} \Delta = 46$ MHz. Thus, the range of observable modulation frequencies in the temporal interferogram produced by mixing mode-locked pulses from two Ti:Sapphire lasers is 46 – 62 MHz.

The minimum modulation period is 16 ns. The maximum modulation period is 22 ns. Figure 9 presents both of these limits. The dashed envelop function shows the modulation waveform “sampled” by the mode-locked pulses. Using a detector with ~60 MHz bandwidth, the individual pulses would not be observed but would instead be smeared out into the envelop function.

![Figure 9 Limits of Pulse Modulation](image)

We can examine the minimum and maximum modulation frequencies from the experiment conducted by Schliesser.\textsuperscript{6} Given pulse width of 10 fs, $f_r = 125.130$ MHz, $\Delta = 232$ Hz, $N_{\text{min}} = 200000$, $N_{\text{max}} = 270000$.\textsuperscript{6} We can approximate that pulses occur every 8 ns within the period of overlap as otherwise there is no interference and no useful modulation. The minimum
modulation frequency is 46.40 MHz with a period of ~22 ns Figure 10 (a) as calculated directly from the $N_{\text{min}}$ and $N_{\text{max}}$. The maximum modulation frequency is 62.64 MHz with a period of ~16 ns Figure 10b. now taking the difference between periods of the roundtrip frequencies $f_r$ and $f_{r+\Delta}$ we calculate 14.8 fs. Dividing the pulse width by this difference in period gives the number of pulses per interferogram as $0.675^6$.

Figure 10 2 Mode and 70 Mode Modulation Simulation
(a) superposition of min and max modulation periods (b) superposition of 70 interference terms

There have only been two components minimum and maximum of the modulation calculated by superimposing them, we see the pulse modulation becomes more varied Figure 10a as compared to those in Figure 9. Now for a larger number of modulation terms taking the 70000 Schliesser says will make up the full modulation spectrum this number is too large to simulate we instead superimpose 70 creating the interferograms Figure 10b.$^6$

Keilmann’s work on Ti:Sapphire generated interferograms informs the following sections goals. Figure 11 shows a micro-second scale interferogram with a kHz frequency spectrum. The radio frequency down conversion factor necessary provided by Keilmann S=43500000 allows
the extraction of infrared information from the radio frequency signal recorded. We will undertake the same process with the goal of obtaining a radio frequency scaled spectrum then conversion to in our case the visible spectrum initially.¹

![Keilmann’s Interferogram and Spectrum](attachment:Keilmann_interferogram_spectrum.png)

**Figure 11** Keilmann’s Interferogram and Spectrum  
(a) Interferogram (b) Spectrum  

With successful beam combining Figure 12 shows the digital oscilloscope trace of the overlapping mode-locked laser pulses with repetition-rate difference (beat frequency) of 12.5 Hz. The signal was collected using an InGaAs detector. Figure 12a presents the total signal with temporally resolved dual beam mode-locked pulses at repetition rate 124 MHz. When the pulses from the two lasers overlap, there is a peak, which defines the centerburst of the interferogram.
On either side, the mode-locked pulses have drifted apart, so that they no-longer temporally overlap or interfere. This region defines a baseline or background signal level. An electronic low-pass 62 MHz filter removes the individual pulses and leaves just the envelop of the interferogram, and a flat line representing the background, as presented in Figure 12b.

![Figure 12 Unfiltered and Filtered Experimental Interferogram](a) Unfiltered 12.5 Hz beat frequency signal (b) 62 MHz Low pass filtered signal

The signal-to-noise ratio for the interferogram in Figure 12b is outstanding. However, there is the possibility that the interferogram is distorted due to detector saturation. This has been resolved by a process described in the Appendix in which an experiment is conducted on the InGaAs detector demonstrating a linear response and no detector saturation distorting the interferogram.

The interferogram in Figure 12(b) was Fourier transformed in produce a spectrum presented in Figure 13. The lower axis scale is the down-converted radio frequencies in the interferogram ranging from 30 to 50 MHz. The upper frequency limit is less than half the 62
MHz folding frequency determined by the pulse rep rate and the Nyquist (sampling) theorem. Modulation frequencies in the interferogram above 62 MHz were already removed by electronic filtering, as in Figure 12b, before the Fourier transform. Converting the rf scale to wavelength results in the non-linear scale given on the top axis of Figure 13. The peak of the spectrum is at ~760 nm wavelength, which agrees with the known emission spectrum of the mode-locked Ti:Sapphire laser.

![Scaled Wavelength (nm)](image)

**Figure 13**: Ti:Sapphire Interferogram Spectrum

Returning to the carrier envelope offset in the context of the interferograms suppose $f_o$ varies independently for the two lasers of a dual frequency comb FTIR, then the effective beat frequency $\Delta_{\text{eff}}$ will vary in time. This is easy to see from the following simplified example. If each laser contains just one mode $f_1 = f_r$ and $f_2 = f_r' + f_o = f_r + \Delta + f_o$, then the effective beat
frequency would be \( \Delta_{\text{eff}} = f_2 - f_1 = \Delta + f_o \), where \( \Delta \) is the nominal beat frequency. If \( f_o \) depends on time, then the temporal positions of the maxima in the interferogram will vary in time.

The negative effect of a time-varying \( f_o \) is that appearance of interferograms on the digitized transient recorded by the oscilloscope will be aperiodic. This would make it difficult to average multiple interferograms, which is necessary to obtain adequate signal-to-noise ratio.

While we cannot measure \( f_o \) directly we can measure the difference between the \( f_o \) of each laser as that offsets the spectrum by a frequency of \( \delta f_o \) which we will refer to as the differential carrier envelope offset. To examine this value an experiment was conducted where a known Nd:S-VAP was placed in the beam so that the absorption lines could be used to offset the resulting spectrum to the known values.

Figure 14 No Absorbing Medium Background and ND:S-VAP Absorbing Laser Crystal

The known values of the two absorption lines were 12350 1/cm and 12517 1/cm. The measured spectrum then showed lines at 12299 1/cm and 12466 1/cm. These are shown in Figure 15 superimposed.
The difference between the calibrated values and the experimental values is 51 1/cm = 616 kHz.

We will now define $\delta f_o$ as $\delta f_o = 616$ kHz for our efforts to obtain the optical frequency.

![Graph](image)

Figure 15 ND:S-VAP

To fully recover the optical frequencies from the radio frequency spectrum we must employ $S = \frac{f_r}{\Delta}$ the frequency down conversion factor along with a more complex radio frequency term to account for the complications of the actual experiment this takes the form of Equation 13.

$$\nu_{opt} = f_o + \frac{f_r}{\Delta} (k f_r + \delta f_o \pm f_{rf}) \quad (13)$$

For the Ti:Sapphire lasers of the experiment we can ignore $f_o$ as it is less than half $f_r$ and is on the order of MHz which is negligible in comparison to the second which is of the order of 100 THz.
The differential carrier offset however is of significant magnitude $\delta f_o = 616 \text{ kHz}$ on order of 1%. The coefficient $k$ represents the order of the spectra with odd and even integers paired to $-$ and $+$ respectively signs on the radio frequency so we define it as $k = (n, \pm)$ in Equation 13. Therefore, orientation of the spectrum is dependent on this sign and integer pairing. It is necessary though to determine the initial $k$ value and sign of the radio frequency from examination of a low enough beat frequency we can be sure is a $k = (1, +)$ order spectra. Figure 16 shows the progression from a 0th order to 1st order $k$. The transition between these two orders occurs when the full spectral radio frequency range exceeds the folding frequency 62 MHz. We also find frequencies where the mirror and true spectrum overlap to be indecipherable as distinguishing one spectrum from the other is impossible Figure 16c demonstrates this difficulty very clearly.
Beat frequency (a) 15.625 Hz (b) 18.75 Hz (c) 21.875 Hz (d) 25 Hz (e) 28.125 Hz

With the necessary values we can now use Equation 13 to scale our directly measured radio frequency properly and consistently to the optical frequencies we are examining. Figure 17 shows the signals already shown in Figure 16a and Figure 16e but now scaled and oriented to the same frequency axis. Their amplitudes are also normalized as in transmittance measurements the amplitude scaling divides out. The spectra should be identical other than a higher resolution at higher beat frequencies. We note however that there is an offset between these two spectra at values below ~385 THz, this remains to be resolved and is under investigation.
Figure 17 Optically Scaled Spectra
Blue 15.625 Hz and Red 28.125 Hz
CHAPTER THREE: DIFFERENCE FREQUENCY GENERATION

For the generation of the infrared spectrum a nonlinear Gallium Selenide (GaSe) crystal is employed. Difference Frequency Generation (DFG) is highly angle dependent and so the critical problem is to simultaneously phase match the various difference frequency generated wavelengths to the optical wavelengths. Using phase matching the efficiency for a given angle can be found.³

The general DFG set up is shown Figure 18. The polarized visible spectrum pulse train is focused by an off axis parabolic mirror of 25 mm focal length. The angle of the GaSe is 70 deg which is the maximum obtainable experimentally. With successful DFG the beam will have a visible and infrared component that travel parallel along the beam path. A germanium filter of 3 mm is employed to filter the visible wavelengths from the beam before the beam is incident on the detector.
The relevant frequencies for DFG are defined in Figure 19. There is a CW spike at 437.5 THz this has not added any meaningful interference to the project at the moment although work is being done to mitigate it.
Figure 19 Ocean Optics Measured Spectrum from Twin Superimposed Ti:Sapphire
(a) $\omega=337$ THz (b) $\omega=385$ THz (c) $\omega=441$ THz

From conservation of momentum and energy we start assuming the mode-locked pulses have a defined spectrum. Conservation of energy is a result of taking $\omega_1$ as the minimum frequency and $\omega_3$ as the maximum in the pump pulse. Then $\omega_2$ is the resulting IR frequency.

$$\omega_3 = \omega_2 + \omega_1. \quad (14)$$

Conservation of momentum gives a relationship between wavevectors $k_3 = k_1 + k_2$, or using $\omega = ck/n$ we find

$$n_3 \omega_3 = n_2 \omega_2 + n_1 \omega_1. \quad (15)$$

Efficient DFG requires that the IR wave co-propagate with the pump pulse (phase matching), which can be achieved using uniaxial crystals. GaSe is a negative uniaxial crystal $\epsilon_\perp > \epsilon_\parallel$. Wave propagation in uniaxial crystals is described in Landau and Lifshitz vol. 8. Propagation speed of ordinary waves is independent of the angle of propagation relative to the crystal’s optical axis, z. Extraordinary waves propagate at a speed that depends on that angle, and it is necessary to distinguish between ray vectors, which give the direction of energy flux, and wave vectors, which define planes of constant phase.

The type of phase matching that we use is Type I, or “eoo”. We want $\omega_3$ to be an e-wave, i.e. horizontally polarized in the principal section (xz plane). We want $\omega_1$ to be an ordinary wave, i.e. vertically polarized and perpendicular to the principal section (along y). However, the incident beam contains both $\omega_3$ and $\omega_1$. We can satisfy the requirement that the input beam simultaneously has a polarization component in xz plane and a component along y by polarizing the pump at 45 deg to the horizontal. The output $\omega_2$ will be an o-wave, polarized along y. Just the
parallel \parallel \text{ component of } \omega_3 \text{ and just the y component of } \omega_1 \text{ satisfy type I phase matching for DFG.}^3

We use thin crystalline plates of GaSe, which are cut so they are perpendicular to the optical axis (z-cut). The crystals are mounted with the z-axis horizontal so that they may be rotated about the vertical y-axis. The x-z plane defines the crystals principal section. The polarization of e-waves is such that their electric field lies in the principal section, and for our experiment, this polarization is horizontal Figure 20. For o-waves, the electric field is perpendicular to the principal section, i.e. vertical, or along the y-axis Figure 20.

![Diagram](image)

Figure 20 z-cut GaSe
(a) e-wave (b) o-wave

The o-wave ray- and wave-vectors coincide, and the refracted beam propagates at an angle \( \varphi' \) is given in terms of the incident beam angle \( \theta \) by Snell’s law

\[
\sqrt{\epsilon_{\perp}} \sin \varphi' = \sin \theta.
\]  

(16)

The phase velocity of the o-wave is \( c/\sqrt{\epsilon_{\perp}} \). The e-waves wave vector angle \( \theta \) in the crystal differs from its ray vector angle \( \varphi' \).
Figure 21 Uniaxial Wavevector Angle

*z*-axis perpendicular and *x*-axis parallel refractive indexes where θ is the wavevector angle.

The propagation speed \( c/n \) of the e-wave depends on θ Figure 21 according to the Fresnel equation

\[
\frac{1}{n^2} = \frac{\sin^2 \theta}{\varepsilon_\parallel} + \frac{\cos^2 \theta}{\varepsilon_\perp}.
\] (17)

Thus, the speed of the e-wave can be adjusted by changing its wavevector angle, which is controlled by changing the angle of incidence \( \theta \).\(^8,9\)

The relationship between \( \theta \) and \( \theta \) is found as follows. The e-wave modified Fresnel Equation 17 can be written

\[
\frac{n_z^2}{\varepsilon_\perp} + \frac{n_x^2}{\varepsilon_\parallel} = 1.
\] (18)
The tangential component of the ray vector must be continuous across the GaSe boundary surface, so that internally \( n_x = \sin(\theta) \). Then from (16) we find \( n_z = \sqrt{\epsilon_\perp - (\epsilon_\perp / \epsilon_\parallel) \sin^2(\theta)} \), and taking the ratio of \( n_x \) by \( n_z \) the wave vector angle can be calculated

\[
\tan(\theta) = \frac{n_x}{n_z} = \frac{\sin(\theta)}{\sqrt{\epsilon_\perp - (\epsilon_\perp / \epsilon_\parallel) \sin^2(\theta)}}. \tag{19}
\]

The ray vector follows similarly and is calculated from the direct relation between the wave and ray vectors \( \tan(\theta') = \epsilon_\perp / \epsilon_\parallel \tan(\theta) \) resulting in

\[
\tan(\theta') = \frac{\epsilon_\perp}{\epsilon_\parallel} \frac{n_x}{n_z} = \frac{\sqrt{\epsilon_\perp} \sin(\theta)}{\sqrt{\epsilon_\parallel (\epsilon_\parallel - \epsilon_\parallel \sin^2(\theta))}}. \tag{20}
\]

For completing the calculation for the ordinary and extraordinary waves we will use Kato’s numerical permittivity equations for the perpendicular and parallel permittivity Figure 22.7,9
We will select three wavelengths 680, 780 and 890 and their corresponding permittivity values and use Equations 16, 19, and 20. Figure 23 shows the resulting incident angle and refracted angle for the extraordinary and ordinary waves. The primary concern will be the extraordinary wave vector results as they will be necessary for the calculation of efficient DFG values.
Figure 23 Incidence vs. Refracted Angle in GaSe
(a) extraordinary ray vector (b) ordinary ray and wave vector (c) extraordinary wave vector

Type one phase matching through angle tuning is necessary for efficient generation and propagation of the DFG spectrum. The manipulation of Equation 15 via substitution of Equations 17 and 19 results in the following

\[
\frac{1}{\sqrt{\epsilon_\perp(\omega_2)} + (\sqrt{\epsilon_\perp(\omega_1)} - \sqrt{\epsilon_\perp(\omega_2)})^2} = \frac{\sin^2 \theta}{\epsilon_\parallel(\omega_3)} + \frac{\cos^2 \theta}{\epsilon_\perp(\omega_3)}. \tag{21}
\]

Substituting in wavelength terms and solving for \(\sqrt{\epsilon_\perp(\omega_2)}\) which will be represented by \(f_R[\lambda_3, \theta; \lambda_1]\), results in

\[
f_r[\lambda_3, \theta; \lambda_1] = \sqrt{\epsilon_\perp(\lambda_2)} = \left(\frac{1}{\lambda_3 - \lambda_1}\right) \left[\sqrt{\epsilon_\perp(\lambda_1)}\lambda_3 - \frac{\lambda_1}{\sqrt{\frac{\sin^2 \theta \cos^2 \theta}{\epsilon_\parallel(\lambda_3) + \epsilon_\perp(\lambda_3)}}}\right]. \tag{22}
\]
The $\lambda_1$ and $\lambda_3$ are the near IR wavelengths in the pump pulse 890 nm and 680 nm respectively.

The procedure is now to choose $\lambda_3$ within the pump spectrum, calculate the range of the pump wavelength $\lambda_1 > \lambda_3$ that give

$$2.88 < \lambda_2 < 14 \, \mu m \quad (23)$$

according to the relationship a wavelength converted Equation 14

$$\frac{1}{\lambda_1} = \frac{1}{\lambda_3} - \frac{1}{\lambda_2}. \quad (24)$$

Choose an incidence angle and calculate the wavevector angle $\theta$ for each $\lambda_3$ in the calculation.

Then, for a given $\theta$, we plot $f_R[\lambda_3, \theta; \lambda_1]$ within the range of $\sqrt{\varepsilon_{perp}(\lambda_2)}$ for the desired $\lambda_2$ range as a function of $\lambda_1$ for $890 \, \text{nm} > \lambda_1 > \lambda_3$. The values of $\lambda_1$ for which $f_R$ falls within the vertical limits of the plot determines the $\lambda_2$ values, which are then calculated from Equation 24.

Figure 24 presents plots of $f_R[\lambda_3, \theta; \lambda_1]$ vs $\lambda_1$. The vertical axes in Figure 24 spans the range of $\sqrt{\varepsilon_{perp}(\lambda_2)}$ expected from for $2.88 < \lambda_2 < 14 \, \mu m$. Each different segment distinguished by different colored symbols corresponds to a different value of $\lambda_3$ as indicated in the legend in $\mu m$. Each separate plot within the figure corresponds to a different incidence angle. Starting from the top, these angles are 70, 65, 60, 55, 50, 45 deg. The horizontal span of each colored curve gives the range of $\lambda_1$ for the considered $\lambda_3$ and $\theta$ that give $\lambda_2$ in the desired range.

Then, the actual value of $\lambda_2$ is calculated from $1/\lambda_2 = 1/\lambda_3 - 1/\lambda_1$ for each $\theta$.

One sees in Figure 24, that different curves have different slopes. The lower the slope, the greater the width of the $\lambda_2$ range for the given $\lambda_3$. Additionally, for a given $\lambda_3$, the wavelengths $\lambda_1$ are longer for the larger $\theta$, giving longer wavelengths $\lambda_2$. The more different curves that
appear in each plot, the more of the pump pulse spectrum that can be used to generate IR, so we expect DFG efficiency to be higher at larger angles.

Figure 24 $f_{2}(\lambda_2, \theta; \lambda_1)$ vs $\lambda_1$. The legend gives $\lambda_3$ values. The plots from top to bottom are for incident angles 70, 65, 60, 55, 50, 45 deg.

Figure 25 presents the calculated values for the infrared wavelength $\lambda_2$ as a function of incidence angle. The IR wavelengths are shorter at the smallest incidence angle, as expected. The values are limited in resolution by the wavelengths selected and angle increments, i.e. due to numerical noise.
Refining the previous method, we can reduce the numerical noise and increase the resolution. Previously established the incidence angle with respect to the normal (z-axis) of the GaSe plate is not the same as the internal wavevector angle \( \theta \) in Equation 22, and it is not trivially determined by Snell’s law. Instead it is found from Equation 19 we selected a range of incidence angles of 45 to 70 degrees. We chose angle steps of 1 deg resulting in 25 separate calculations. The pump wavelength range was 680 to 890 nm, and this range was divided into 1000 points to give 0.21 nm resolution. The spectral limits of the pump define the minimum
possible DFG wavelength to be 2.88 μm. The maximum DFG wavelength of interest was
decided to be 14 μm. These two limits determine limiting values for the left side of Equation 22
based on the empirical values of $\varepsilon_\perp$. Equation 23. The shorter $\lambda_2$ limit corresponds to the larger
limit in Equation 23. For a given angle of incidence, Equations 22 and 19 give solutions $\lambda_2$ that
span only part of the considered range 2.88 μm < $\lambda_2$ < 14 μm. We next describe how the upper
and lower $\lambda_2$ limits of the solution to Equation 22 are determined at each angle of incidence.

The right side of Equation 22 we again call $f_R[\lambda_3, \theta; \lambda_1,]$. The method of solution is to
choose an incidence angle $\theta$ and the shortest wavelength $\lambda_3$ within the pump beam. Then we plot
$f_R$ as a function of $\lambda_1$ for all $\lambda_1$ within the pump beam that satisfy $\lambda_1 > \lambda_3$. We limit the range of
the plot to the values defined by Equation 23. Then we choose a longer wavelength $\lambda_3$ within the
pump beam and generate a new curve. The process is repeated until the curves no longer fall
within the limits of the plot.

Figure 26 presents such a plot 70 deg incidence angle. One sees that the curve for $\lambda_3 =
0.8$ μm is the one with the longest $\lambda_3$ value that still fits within the plot, and it does so at the
longest possible $\lambda_1$ value near 0.89 um wavelength. We then look for the slightly longer $\lambda_3$ value
that gives exactly $f_R = 2.671$ when $\lambda_1 = 0.89$ um, i.e., where the $f_R$ curve is given by a single point
in the lower right corner of Figure 26. Then the corresponding DFG IR wavelength $\lambda_2$ is found
according to Equation 24. According to that procedure we find a $\lambda_2$ value near 7.9 μm. The
procedure just described determines the lower bound on the DFG IR wavelength for the given
incidence angle. The procedure to find this lower bound is repeated for the different incident
angles considered.
The longest $\lambda_2$ value will be determined by the shortest $\lambda_1$ for the shortest $\lambda_3$ in the pump beam, which is always 0.68 $\mu$m. When the $f_R$ curve for $\lambda_3 = 0.68$ $\mu$m extends from bottom to top of the $f_R$ plot, as for the curve with black square symbols in Figure 26, then the shortest $\lambda_1$ can be found by extrapolating the curve joining the symbols to the bottom horizontal axis. Or the resolution of $\lambda_1$ points can be increased until a point on the curve coincides with the lower border of the $f_R$ plot. The corresponding $\lambda_1$ value gives the longest possible $\lambda_2$ value. For instance, from Figure 26, the line through the black square symbols intersects the bottom axis at $\lambda_1 = 0.719$ $\mu$m, and then Equation 24 gives $\lambda_2 = 12.54$ $\mu$m.

To summarize this calculation, we have looked at if 680 nm is assumed what wavelengths longer than it couple to produce DFG IR values and 890 nm is examined for what
values shorter when coupled with it produce DFG IR. This does not represent all values that can be coupled but only those that satisfy the conditions described. Figure 27 presents the upper (680 nm) and lower (890 nm) DFG IR values calculated. The plot is truncated at 14 μm, which means only that we have no interest in longer wavelengths. It does not mean that such wavelengths are not generated at incident angles below 60 deg. A MCT detector has been selected that is sensitive from 830 nm to 16 μm. We note that in Figure 27 from experimental data and this calculation we can expect to see 10 μm at incidence angles above 55 degrees. This begins to align with the centerline by Brehm and while there is some deviation, we can see that the curve of our calculated line is nearly identical and is offset linearly in comparison to the data.¹⁰

Figure 27 Experimental and Theoretical DFG Values
We will expect the power of the infrared signal when the visible red frequencies have been filtered out through Germanium filters to be on the order of \( \sim 10 \mu W \).\(^6\) Interferograms in the infrared are anticipated were beating of the infrared beams will be achieved using a thin ZnSe beam combiner and wedge to preserve optical path length. We will also be able examine in great deal the relationship between incident angle and resulting spectrum. The calculation described can also be further refined to give a more complete model of the DFG available wavelengths. This is where the current experimental efforts are being focused as these remain the most critical issue to be resolved. Once real data from our experiment has been obtained, we can begin packaging data to be sent to the AI and begin generating the library of spectral information of various pathogens for the onsite spectral instruments.
APPENDIX: LINEARITY OF INGAAS DETECTOR
To confirm that the visible wavelength interferograms generated in this experiment are not distorted due to detector saturation an experiment was conducted for an InGaAs detector. First, we determine whether the Neutral Density (ND) filter follows the manufacturer specification. Figure 28 a plot of the neutral density filter attenuation specified by the manufacturer. The data (smooth curve) are plotted as transmission vs optical density (OD), $T = 10^{-OD}$. The symbols in Figure 1 are our measurements of transmitted laser signal measured with the Si power meter as a function of ND filter segment. The upper limit on the power meter is 500 mW, so that we may assume its response is linear in the 0-4 mW range of the experiment. Comparison of the two curves shows that the attenuation of the ND filter increases more rapidly than specified.

![Figure 28 Theoretical ND Filter Compared to Measured Power](image)

*Figure 28 Theoretical ND Filter Compared to Measured Power*  
(smooth curve) Specified transmittance of variable 8-segment ND filter. (symbols) Attenuated laser power measured with Si power meter.
Next, we addressed the concern that saturation of the Thorlabs InGaAs DET10N2 was suppressing the peak of the preliminary interferograms obtained from the combined mode-locked laser beams. To test this, the combined beams were attenuated by the variable ND filter. An interferogram was recorded along with the non-interfering background of ~124 MHz mode-locked pulses that had drifted out of synchronization, so that they no longer overlapped. The interferogram peak and average background (symbols) are compared in Figure 29 to expectations based on the ND filter specifications (smooth curves). The disagreement between symbols and curves is comparable to what was reported in Figure 30 for the linear Si power meter.

![Graph showing interferogram and background compared to expected ND filter](image)

**Figure 29 Interferogram and Background Compared to Expected ND Filter**

Interferogram peak and background (symbols) measured with InGaAs detector compared to expected signal based on ND filter specifications (solid curves).

The ratio of interferogram peak to background is plotted in Figure 30. The legend indicates the ND filter segment used. The dependence is linear within the experimental uncertainty. This indicates that the InGaAs detector is not saturated at the peak of the interferogram centerburst, so that the interferogram should not be distorted.
Figure 30 Interferogram vs. Background
Average signal of non-overlapping pulses vs peak interferogram signal from region of overlapping pulses. The different points correspond to different attenuation levels given by the ND filter segment indicated in the legend.
REFERENCES


