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Dual-Etalon, Cavity-Ring-Down, Frequency Comb Spectroscopy

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Abstract

The ”dual etalon frequency comb spectrometer” is a novel low cost spectometer with limited moving parts. A broad band light source (pulsed laser, LED, lamp ...) is split into two beam paths. One travels through an etalon and a sample gas, while the second arm is just an etalon cavity, and the two beams are recombined onto a single detector. If the free spectral ranges (FSR) of the two cavities are not identical, the intensity pattern at the detector with consist of a series of heterodyne frequencies. Each mode out of the sample arm etalon with have a unique frequency in RF (radio-frequency) range, where modern electronics can easily record the signals. By monitoring these RF beat frequencies we can then determine when an optical frequencies is absorbed. The resolution is set by the FSR of the cavity, typically ¡ 10 MHz, with a bandwidth up to 100s of cm^{1}. In this report, the new spectrometer is described in detail and demonstration experiments on Iodine absorption are carried out. Further we discuss powerful potential next generation steps to developing this into a point sensor for monitoring combustion by-products, environmental pollutants, and warfare agents.

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Summary

Absorption spectroscopy is the most quantitative and straight forward means of measuring the concentration and temperature of a species having a known spectrum. In order to do this quantitatively one needs to perform the absorption measurement with high resolution over a large bandwidth. With high resolution one can measure Doppler profiles to obtain temperature and extract accurate absorption measurements at the center of an absorption feature to determine concentration. With broad bandwidth one can obtain rotational and vibrational distributions that can be fit to temperatures as well as monitor several species at once. We describe a simple spectrometer that will allow one to do both high resolution and broad bandwidth simultaneously and the measurement requires only a few microseconds requiring no special light source or electronics.

The spectrometer relies upon a beam from a broad bandwidth light source (laser or CW, diode or lamp), that can be pulsed or continuous, that is split into two beams. One beam is directed through a confocal etalon having a free spectral range (FSR) of the required spectroscopy. This etalon creates a frequency comb. This light of this beam is also directed through an absorbing medium. The absorber can be before, after or inside of the confocal etalon. If the absorber is placed inside the etalon this is a cavity ring Down arrangement and one obtains greatly enhanced sensitivity. The other light beam (the Read Out beam) is directed through a separate confocal etalon and is not subject to the absorber. The read out beam can either be a single frequency (obtained by using a short etalon with a large FSR) or multiple frequencies that are similar but not quite the same as the frequencies of the beam coming from the absorption arm of the spectrometer. By careful picking of the frequencies in the read out arm of the spectrometer and combining these beams on a photodiode and recording the interference pattern obtained from the cross beat frequencies one can reconstitute the absorption spectrum. This absorption spectrum will comprise a series of discrete absorption measurements with the spacing of the FSR of the absorption arm confocal etalon each with the resolution associated with the convolution of the finesse of the two confocal etalons.

In order to obtain 1MHz resolution of the individual absorption measurements requires a ring down time of the etalons of approximately 1 microsecond. If that interferogram is analyzed in parts, every 100 nanoseconds for instance, then one can obtain a time-resolved spectrum with about 10 MHz spectral resolution and 100 nanoseconds time resolution. In order to obtain 1 MHz spectral resolution with a traditional Michelson interferometer type of Fourier Transform spectrometer would require moving the mirror a kilometer distance. Additionally, in order to obtain the 100 nanosecond time-resolution would require hours of operation in a step scan mode. Our measurement is 7 orders of magnitude faster.

1 Introduction

Absorption spectroscopy is one of the most quantitative methods for identifying unknown gases, liquids, and aerosols in either the laboratory or real world applications. For instance, in the infrared portion of the spectrum, many molecules contain a “fingerprint”, a unique pattern of optical transitions that are distinct due to the compounds chemical moiety (OH, CH, CO). The primary issues with using absorption spectroscopy as an everyday tool for identifying environmental contaminates are that large spectral bandwidths with good resolution are needed and high sensitivity is required. Typically contaminates of the greatest concern/interest are very dilute in the environment so some manner of increasing the sensitivity of direct absorption methods are needed for sensor development. The most obvious manner for obtaining enhanced sensitivity is to multipass the light to obtain a longer path length for the absorption. This can take the form of a cavity ring down cell or a simple multipass design [4, 9, 7, 8].

Cavity-ring-down based spectrometers, which use optical resonators to increase the effective path length of the absorbing medium, greatly improve absorption sensitivity to dilute substances are now routinely used to monitor green house gasses. However they are generally limited to measuring absorption at extremely narrow spectral regions associated with the laser light source used. Modern day state-of-the-art spectrometers attempt to overcome the issues of spectral resolution (selectivity) at the cost of bandwidth. Generally, high-resolution laser light sources are used that are resonant with a atomic or molecular transition of interest, but these sources are not broadly tunable. When using a broad light source the frequency resolution is limited by the spectrometer [6, 12, 11] .

Here we demonstrate a new absorption based spectrometer capable of both high sensitivity and broad bandwidth spectral coverage with high spectral resolution. This spectrometer is inspired by recent laboratory experiments that have demonstrated a spectrometer that uses two phase-locked femtosecond lasers (Frequency Comb Lasers) in conjunction with a cavity ring-down-cell in order to obtain broadband absorption spectroscopy with the enhanced sensitivity necessary to detect dilute species[12, 10, 2, 5, 1].

The novel spectrometer described here has the large bandwidth needed for chemical fingerprinting, superior resolution to current commercial spectrometers and the sensitivity of cavity ring down spectroscopy but, in principle, no moving parts or high spectral resolution lasers systems are needed. With the design of this spectrometer it is straight forward to engineer it into a sensitive point sensor for detection of air born pollutants and toxins. We report the first demonstration of its operation. The “Dual-Etalon, frequency - comb, cavity-ring-down spectrometer ” uses one light source (a laser, LED, lamp,) and two air-gap Fabry-Perot etalons to imprint two different frequency patterns onto the broad band light source, creating two transient frequency combs (the lifetimes of which is dictated by the ring down time of the etalon cavities). The two etalons have different frequency outputs and this difference is what allows us to obtain the high spectral resolution. The spectrometer is comprised of two optical paths each containing an etalon. One etalon is used as the absorption arm, while the second etalon is used to “read out” the optical frequencies transmitted by the first arm. The beams coming from the two arms of the spectrometer are combined using fiber optics and the combined signal is directed onto a fast photodiode. Interference between the different frequencies present in the two arms of the spectrometer creates cross beat frequencies in the photodiode response. The cross beat frequencies between the frequencies in the absorption arm with those in the readout arm of the spectrometer are filtered and recorded. As a frequency in the absorption arm disappears due to absorption by an absorbing medium then the cross beat frequencies associated with that frequency will also disappear. It can be arranged, by selection of the etalon lengths, that the cross beat frequencies fall in the hundred to thousands of hertz range so that standard detectors and electronics can record and analyze the beat pattern. By analyzing portions of the beat pattern separately time-resolved spectroscopy can be obtained.

The absorbing medium can be either before, after, or within the confocal etalon. As the technique relies on the use of etalons, a version of cavity-ring-down spectroscopy can be performed if the sample of interest has either weak absorptions or has low concentration. In this case the sample is placed inside the absorption arm etalon and experiences many passes of the light. This multi-pass absorption technique effectively increases the sensitivity of the spectrometer nearly four orders of magnitude.

2 Background

In order to make a quantitative measurement of species by measurement of its absorption it is important to make the measurement with enough resolution to extract the absorbance at the center of the absorption line. One must also know the temperature and pressure of the system as these will change the absorption spectrum. If one can measure the spectrum of a molecule with sufficient resolution over a sufficient frequency range then one can extract the temperature from the spectrum as well as measure the pressure broadening associated with collisional processes. Most absorption based techniques rely upon either a narrow band light source whose frequency can be scanned over the absorption spectrum or a broadband light source whose transmittance can be resolved by a spectrometer. The spectrometer can be grating based such that it relies upon dispersing the light onto a position sensitive detector (or scanned across a slit) or a Fourier Transform based detection scheme such that it relies upon interference of the transmitted light with light that was not put through the absorption cell. As certain frequencies are absorbed the interference pattern changes. In this report we present a new design for a Fourier-transform-based spectrometer capable of measuring megahertz resolution spectra over many hundreds of thousands of megahertz during a single laser shot lasting a few microseconds. This spectrometer is inspired by spectroscopic techniques recently developed using femtosecond-pulse-length, frequency-comb lasers [9, 6].

Over the last decade the advent of frequency-comb lasers has ushered in a new set of tools to use in order to optically identify a specific molecule. A frequency-comb laser is a train of femtosecond laser pulses that are phase locked. The Fourier transform of the train of pulses is a set of frequencies separated by the pulse separation frequency. The frequency width of each tooth (frequency) is given by the Fourier transform of the duration of the coherence of the pulse train. For instance a pulse train that has a repetition rate frequency of 100 MHz and remains phase locked for 1 second will have a set of frequencies (teeth) that are 100 MHz separated from each other, each with a frequency width of 1 Hertz. Any one pulse from the femtosecond laser will not exhibit frequency modulation only the train of pulses carries the frequency modulation of the pulse separation and this modulation is called a frequency comb.

The availability of frequency-comb lasers has recently led to new spectroscopic techniques [11, 10, 2]. For instance if a broadband femtosecond frequency-comb laser is passed through an absorbing sample some of the “teeth” of the frequency comb will be absorbed and others won’t. The question is how does one quantitatively determine which teeth were absorbed. One method for observing the comb teeth of a broad band laser beam that has passed through an absorptive media is to combine the broadband laser output light with a single frequency laser on a fast detector (photodiode) (10). Heterodyne beat notes will be observed at the difference frequencies between the single-frequency laser and each of the comb teeth. A Fourier transform of the heterodyne signal will reveal the frequency comb and any absorption. This technique is limited to beat frequencies that the fast photodiode can respond to and the digitizing speed of the signal acquisition device, about 15 GHz. As 15 GHz is equivalent to half a wavenumber of frequency range the high-resolution spectra can only be obtained for this modest frequency spread using a (non-tunable) single frequency laser to read out the comb teeth. This is very similar in concept to the proof of principle experiment that we report here. However, the spectrometer reported here does not require the use of any frequency comb lasers.

A second technique (multi-heterodyne spectroscopy) has been developed to get around the need for fast photodiode detectors and expensive digitizers. In order to simultaneously recover the absorption information over a much broader frequency range Schiller [10] in 2002 proposed a technique that utilized a second frequency comb laser with slightly different repletion rate to “readout” the absorption information encoded in the first frequency comb laser. This is accomplished by superimposing the two frequency-comb laser beams on a detector and because the frequencies were only slightly different (assume 100 Hz for sake of this example) one could induce beat frequencies between each of the frequency “teeth” in the combs of the two laser beams. If the first tooth in the frequency bomb overlapped in frequency with the first tooth of the second frequency comb then the second tooth of each comb is separated in frequency by 100Hz, the third tooth of each comb is separated by 200 Hz and so on. By recording the low frequency beat notes obtained by heterodyning of the two laser beams the absorption spectrum can be extracted from underneath the frequency bandwidth of the femtosecond laser. This idea was successfully demonstrated by the group of Picque and Hansch [2] in 2009. In this paper they used two frequency comb laser separated in frequency by 100 Hz. One frequency-comb laser beam was injected into a cavity-ring-down cell with its mode spacing matching that of the frequency comb laser. Either ammonia or acetylene was put into the ring-down cavity and the vibrational overtone spectra recorded. The other frequency-comb laser beam was directed around the cell and the two beams combined on a photodiode. The Fourier transform of the signal revealed the absorption of the gas in the region around 800 nm to be encoded between 100 Hz and 100KHz (the vernier-like cross-beat frequencies). In this manner they were able to record the spectrum of acetylene over 250 cm^{-1} (7,500 GHz) bandwidth, with approximately one GHz resolution, in only 40 microseconds. Our Dual Etalon technique is capable of performing this same sort of broadband spectral absorption measurement with the same sensitivity and spectral resolution.

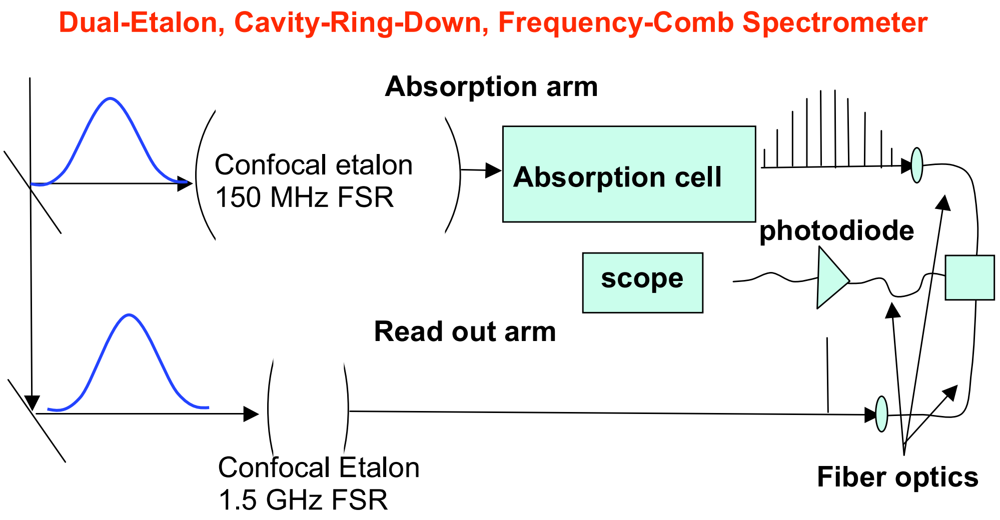

Figure 1. Schematic of dual-etalon cavity-ring-down frequency-comb spectrometer. A light pulse is split and put through two arms of a spectrometer. One contains an absorbing medium and a long confocal etalon (absorption arm). The other arm contains only a short confocal etalon (read out arm). The outputs of the two arms are combined using fiber optics and illuminate a fast photodiode. The current from the photodiode is recorded on a digitizing scope and Fourier transformed.

3 Dual-Etalon, Frequency-Comb Spectrometer

In figure 1 is a schematic of the Dual-Etalon, Cavity-Ring-Down, Frequency-Comb spectrometer. A single laser pulse is split into two pulses. One travels down the absorption arm of the spectrometer. The absorption arm contains a confocal etalon with a free spectral range of 150 MHz and a cell with an absorbing gas. If the gas is put inside the confocal etalon then one is performing multi-pass cavity-ring-down spectroscopy. If the gas is in a separate cell either before or after the confocal etalon then direct single or multi-pass absorption can be performed. The second arm of the spectrometer (the read out arm) contains a short (about 1 cm in length) confocal etalon with a Free Spectral range of 1.5 GHz. The outputs of the two arms are independently focused into two, single-mode fiber optic cables. These are combined into a single fiber optic and this combined beam illuminates a 3 GHz silicon photodiode. The signal from the photodiode is recorded on a Tektronics oscilloscope (5 GPS) capable of performing a Fourier transform on the recorded signal. As the laser pulse used as a light source for the experiments described here has a bandwidth of approximately 1.5 GHz approximately 10 frequencies will be transmitted through the 150 MHz absorption arm confocal etalon and only a single frequency will be transmitted through the 1.5GHz read out arm confocal etalon. The resolution associated with each frequency is determined by the mirror reflectivity (ring down time) of the etalon.

In figure 2 are examples of the signals one obtains from each arm of the spectrometer independently and from the combination of beams on the spectrometer. By blocking first the read out arm and letting only the output of the absorption arm of the spectrometer strike the detector a dramatic interference pattern is observed. When Fourier Transformed this pattern reveals a set of equally spaced frequencies separated by the 150 MHz free spectral range of the confocal etalon. When the absorption arm of the spectrometer is blocked and only the read out arm is allowed to strike the detector a smooth decay is observed because only one frequency is exiting the confocal etalon and therefore no heterodyne beat frequencies are observed in the Fourier transform.

When the two light beams from the two arms of the spectrometer are combined on the photodiode a new set of frequencies appear in the Fourier transform. They are the cross-beats caused by the heterodyning of the single frequency from the read-out arm and the multiple frequencies of the absorption arm of the spectrometer. There is one new cross beat frequency for each of the ten frequencies of the light source emanating from the 150MHz confocal etalon in the absorption arm of the spectrometer. The frequencies of the beats are the frequency separation between the individual frequencies carried in the two beams. In figure 3 is a schematic of the frequencies that compose each light beam after they exit the confocal etalons and the frequency pattern one expects if there is no absorbing media in the absorption arm of the spectrometer that would absorb a specific comb tooth.

The optical detector measures the intensity of the incident light, where $I = c\varepsilon_0 \langle E_1 + E_2 \rangle^2/2$, with c being the speed of light, ε_0 is the permittivity of free space, and E is the electric field of the incident light. While each individual beam has its unique time structure do to the shape of E^2, when both arms are incident on the detector the combined intensity pattern is given by :

$$I(x,t) = c\varepsilon_0 \langle E_1 + E_2 \rangle^2/2. \tag{1}$$

This squaring of the total electric field reveals a cross term dependant on both E_1 and E_2, see equation 2 below:

$$I(x,t) = [(E_1E_1^* + E_2E_2^* + E_1E_2^* + E_1^*E_2)] = c\varepsilon_0 \cos[(\pm\kappa_1 \mp \kappa_2)x - (\pm\omega_1 \mp \omega_2)]t, \tag{2}$$

where κ is the wave vector and ω is the frequency of the light. Since the absorption and read out beams both hit the detector at a given x and t, this further simplifies the results, such that the cross term between the beams reduces to

$$I_{cross} = c\varepsilon_0 \cos[\alpha(\pm\kappa_1 \mp \kappa_2)], \tag{3}$$

where α is a constant determined by the physical parameters of the spectrometer and the speed of light. Thus the cross term gives rise to beat frequencies at the difference frequency between the two arms of the spectrometer.

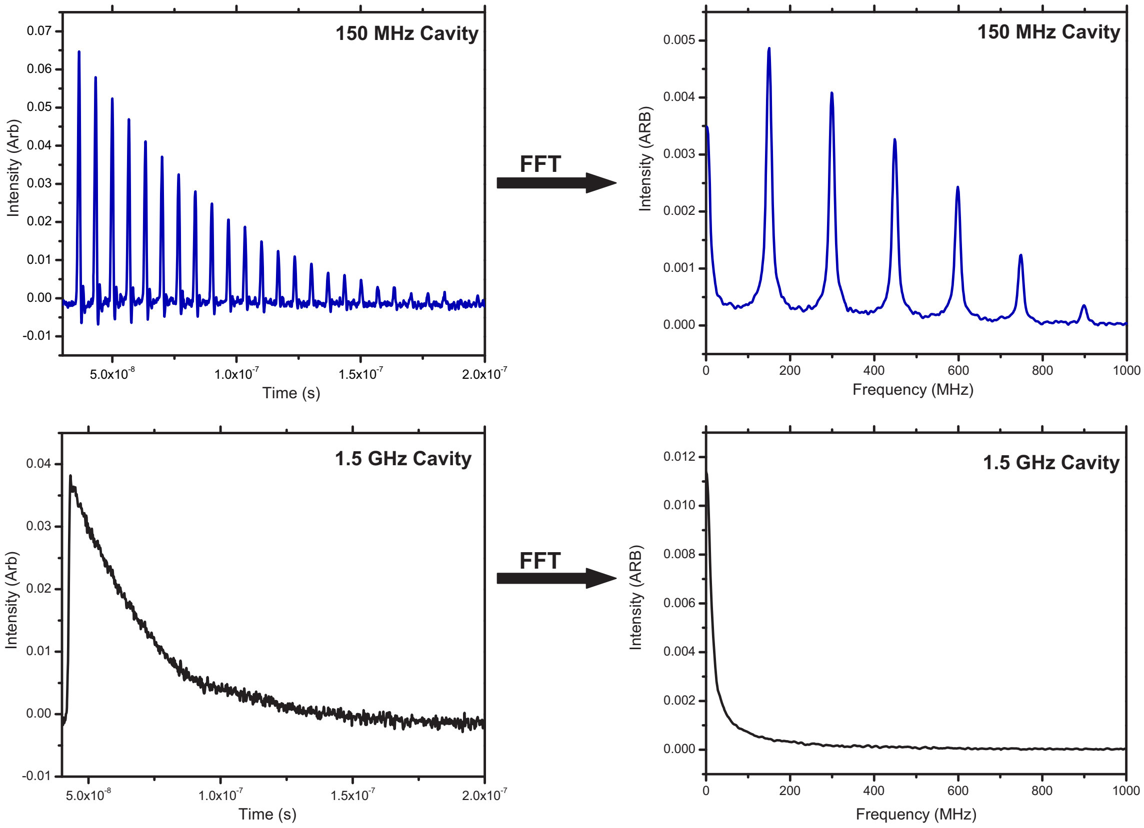

Figure 2. Shown on the top of the figure are the raw signal on the photodiode when a 150 MHz confocal etalon is in the absorption arm of the spectrometer and the Fourier transform of the signal when only the absorption arm of the spectromenter is striking the photodiode. Below are the raw signal and the Fourier transform when a 1.5GHz MHz confocal etalon is in the read out arm of the spectrometer. The light source for both of these signals is the output of a Nd:YAG pumped dye laser (Lambda Physik) that has a bandwidth of approximately 1.5 GHz.

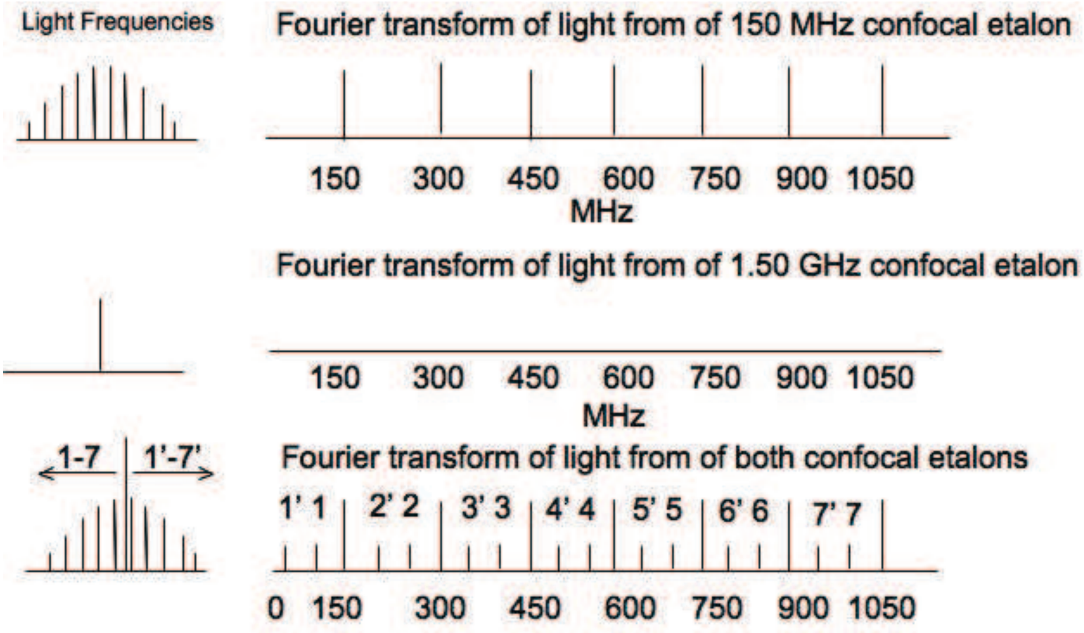

Figure 3. Schematic of the frequencies and the Fourier transforms associated with that frequency structure coming from the absorption arm of the spectrometer (upper panel), the read out arm of the spectrometer (middle panel) and the frequencies expected from the combined light sources (lower panel).

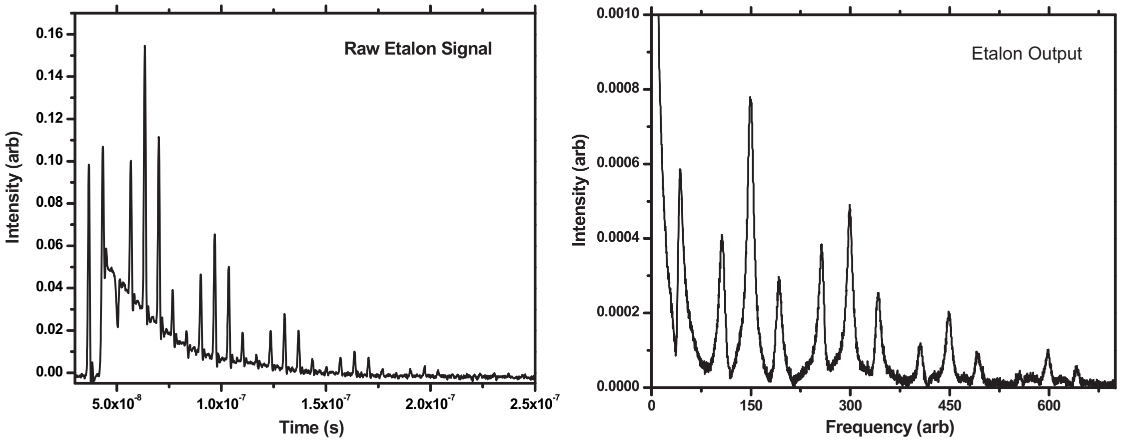

Figure 4. The left plot is raw signal from photodiode when the light beams going down the absorption arm of the spectrometer and the light beam going down the read out arm of the spectrometer are combined on the photodiode. Clear modulation in the interference pattern is observed. The right plot is of the Fourier transform of the raw signal showing the two new beat frequencies between each tooth of the frequency comb of the single absorption arm light (Figure 2).

Figure 4 shows the raw signal of the beat pattern on the photodiode when the two beams are combined. This should be compared to the beat pattern observed when only the absorption arm light is allowed to strike the detector, top of Fig. 2. Also in Fig. 4 is shown the Fourier transform of his photodiode signal showing the cross beats between the two light beams. By comparison with Fig. 2, one can clearly see two extra beats between each comb tooth of the single 150 MHz absorption arm signal, as is schematically predicted in Figure 3. The beat pattern if Figure 4 is obtained because the single frequency light from the small confocal etalon is near the central frequency of the light pulse. As there are as many frequencies from the long confocal etalon associated with the absorption arm of the spectrometer that are to higher frequency as there are to lower frequency we observe a symmetric cross beat pattern. The resolution of the individual light components of the frequency combs is given by the uncertainty broadening associated with the ring down lifetime of the confocal etalon. Because of the approximately 60 nanoseconds decay time of the short confocal etalon we are limited to a frequency bandwidth for the individual beat notes of approximately 20 MHz. This is the ultimate resolution limit of the spectroscopy we are able to obtain with this technique. The commercial confocal etalons used in this experiment are not optimized for long ring down times and the resolution can easily be improved with the use of higher reflection mirrors by orders of magnitude.

In order to obtain a stable beat pattern, both etalons are locked to a stabilized continuous wave (CW) diode laser. The diode laser is locked using an FM Pound-Drever-Hall setup to stabilize the laser to a Doppler free ${}^5S_{\frac{1}{2}} \leftrightarrow {}^5P_{\frac{3}{2}}$ transition in a room temperature Rubidium vapor cell at 780.246 nm. Each etalon cavity was individually locked to the diode laser using an off-set PID (proportional-integral-differential) lock. The etalon cavity length is adjusted via a ring piezoelectric between the Invar cavity and the output mirror.

In this locking technique a DC offset voltage is used to shift the output signal of the laser diode from the etalon such that the signal spans from a negative voltage to positive voltage. The PID circuit then locks the cavity to the zero crossing of the etalon output from the diode laser. By scanning the offset voltage, on either etalon, the length at which the cavity is locked is scanned. If the DC offset voltage on one cavity is adjusted while the other remains constant, the difference frequency between the cross beats can be fine tuned, allowing them to be resolved in the Fourier transform. The sample absorption can be continuously tuned in this manner.

Depending upon the exact frequency coming from the single frequency (1.5 GHZ) etalon with respect to the frequencies associated with the 150 MHZ etalon there is an ambiguity as to ordering of the cross beats underneath the frequency envelope of the light source. That is, in figure 3 do the beats labeled with the prime march to higher or lower frequencies? One can resolve this uncertainty by slightly lengthening either of the confocal etalons, by moving the mirror attached to the piezoelectric, and thereby scanning the stable etalon modes to longer wavelength while one observes the cross beat pattern. In this manner it is possible to determine the absolute ordering of the frequencies underneath the laser bandwidth.

4 Absorption Measurements

In order to demonstrate spectroscopic measurements we placed Iodine vapor into a three-meter long absorption cell in front of the confocal etalon in the absorption arm of the spectrometer. In this manner, the absorption spectra of Iodine is used to demonstrate that we can obtain spectral resolution greater than the laser bandwith of the Sirah dye laser which has a specified resolution of ~ 0.05 cm^{-1} or 1.5 GHz resolution. The laser frequency is scanned until we observe an isolated absorption feature at 16694.5 cm^{-1} energy. In order to observe the shape of the absorption feature in the beat frequencies of the Fourier transform of a single laser pulse we stopped the scan of the laser frequency to be on the edge of the absorption feature. At this frequency some of the teeth should be absorbed and some transmitted and we should be able to record this edge of the absorption by measuring the intensity pattern of the cross beats

As the cross beat frequencies are 20 MHz wide (limited by the Fourier transform of the small etalon decay time) and we can easily obtain with the frequency resolution of our photodiode and oscilloscope 8 resolved beat frequencies representing 1.2GHz of frequency spread at one setting of the etalons and laser (see Figure 3). The beat frequencies that originate from light coincident with an I_2 absorption line will be diminished because the frequency tooth that generated that particular beat note will be diminished. In this manner we obtain spectrum over 1.2 GHz with 150 MHz resolution from a single laser pulse.

To demonstrate this we place our laser on the edge of an absorption band at 16694.5 cm^{-1} (598.996 nm). We obtain a beat pattern with no gas in the absorption cell. This patterns is shown in Figure 5A. We then fill the cell with iodine gas and rerecord the beat pattern again, Figure 5B. We use the beat pattern with no gas in the cell, Fig. 5A, to normalize the beat pattern obtained with the Iodine in the cell, Fig 5B, In Figure 6 are plotted the normalized intensities. In Fig. 6 is plotted the normalized intensity of the individual cross beats from Figure 5B normalized by the intensity of the corresponding cross beasts of Figure 5A. The falling edge of the absorption line is dramatically observed and one can see that the beat frequencies that are near the peak of the absorption have been $\sim$ 80% extinguished. This is to be contrasted to the bulk absorption measurement. When scanning the dye laser over the absorption feature and recoding the transmittance of the light one observes only a transmission minimum of 50% due to the bandwidth of the laser being larger than the absorption feature. That is, when the laser frequency is at line center the wings of the laser are off of the absorption and the measured absorption is not accurate. However as the individual comb teeth of the laser are only 20 MHz wide when one coincides with the much broader absorption feature a quantitative measure of the line-center absorption is obtained.

If one wanted higher resolution then one could scan the length of the etalon in the absorption arm of the spectrometer and scan the individual comb teeth thereby obtaining 20 MHz resolution in the spectroscopy. We note that, in this particular case, one could also scan the length of the 1.5 GHz etalon over the laser bandwidth and use that narrow band light transmitted by the etalon to perform the absorption measurement. However, our objective here is to demonstrate that we can easily observe the cross beats produced by the Dual Etalon spectrometer not obtain another spectrum of Iodine vapor.

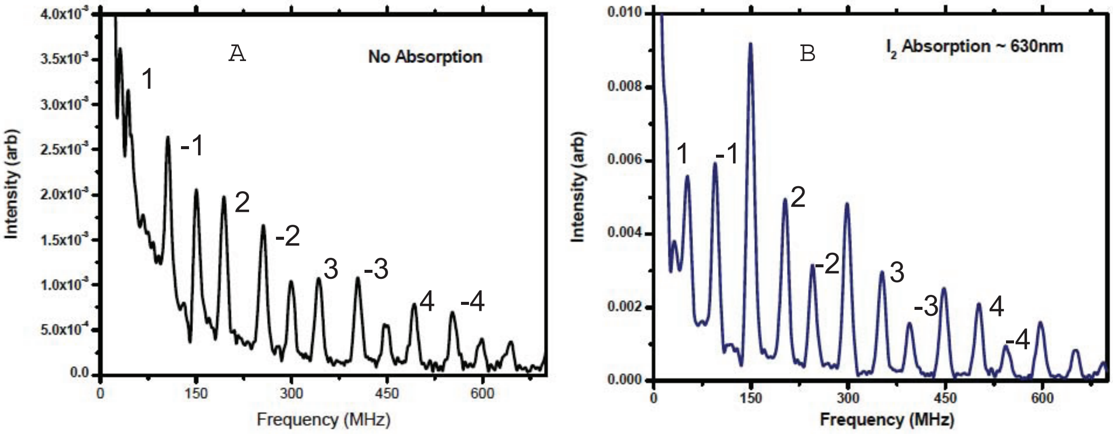

Figure 5. A: the beat pattern observed when there is not absorbing Iodine in the absorption arm of the spectrometer. B: The beat pattern observed when the room temperature vapor pressure of Iodine is introduced into a three-meter long absorption cell in the absorption arm of the spectrometer and the laser is tuned to the edge of an absorption feature.

In another demonstration of the ability to record spectra with the dual-etalon, frequency-comb spectrometer we step scanned the laser across the absorption feature at16694.5 cm^{-1}. As we did this we readjusted the etalon length of the 1.5GHz etalon so that the single frequency tooth of the read-out-arm etalon remained near the peak of the laser light (maximum etalon transmittance) as we step scanned the laser. After we stepped the laser frequency by moving the gratings we re-peaked the amount of light transmitted by the 1.5 GHz etalon by adjusting its cavity length. We recorded the intensities of the cross frequency beats after each step and in figure 7 is a plot of a single beat frequency as the laser scans over the absorption. This is equivalent to step scanning a laser beam with 20MHz resolution over the iodine absorption feature. Also plotted in figure 7 is the transmission profile of the laser light laser through the iodine absorption cell while scanning across the feature as in a traditional absorption measurement (solid line) as well as the transition as it is recorded in the Iodine atlas (triangles). This absorption feature is seen independently of which cross beat frequency one chooses to follow as the laser is step scanned. Once clearly sees the I$_2$ absorption feature but it is not significantly narrower than the feature as scanned by the Sirah dye laser. This is because the feature is inherently about the width of the laser bandwidth and the precision of the step scan of the laser as well as the ability to scan the small etalon to the peak of the laser after each step introduces uncertainty to the frequency position of each point. However this does clearly demonstrate the ability to record absorptions associated with individual comb teeth of the frequency comb and the absorption feature does match well the width as reported in the iodine atlas.

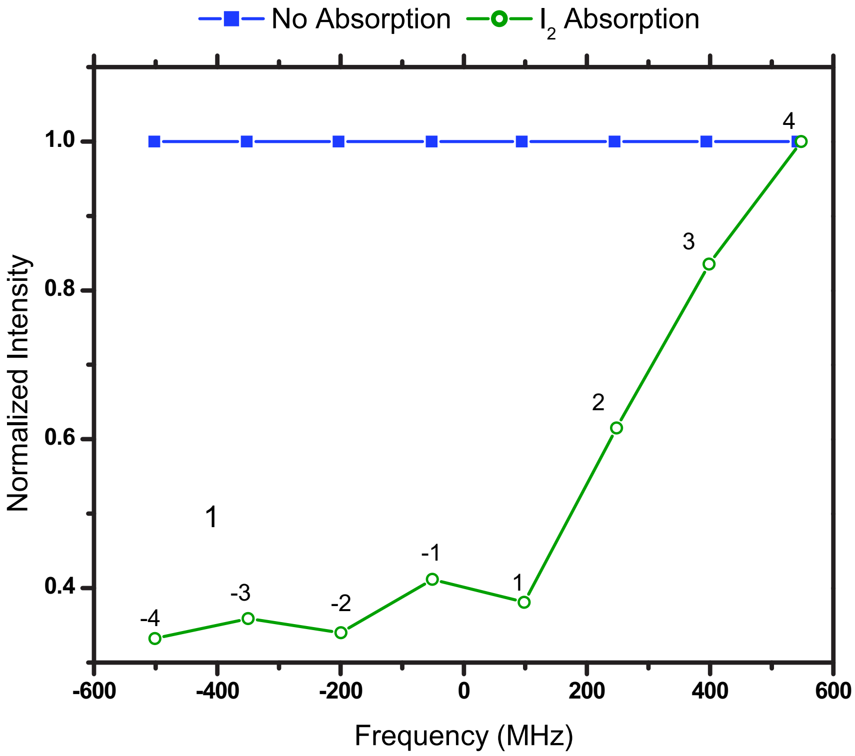

Figure 6. Normalized beat frequency intensities while laser frequency coincides with the edge of the absorption feature at 16694.5 cm^{-1}. The raw beat frequencies are shown in Figure 5A and 5B. The beat frequencies plotted in the order indicated in Figure 5 (4,3,..1,1’,2’3’,4’).

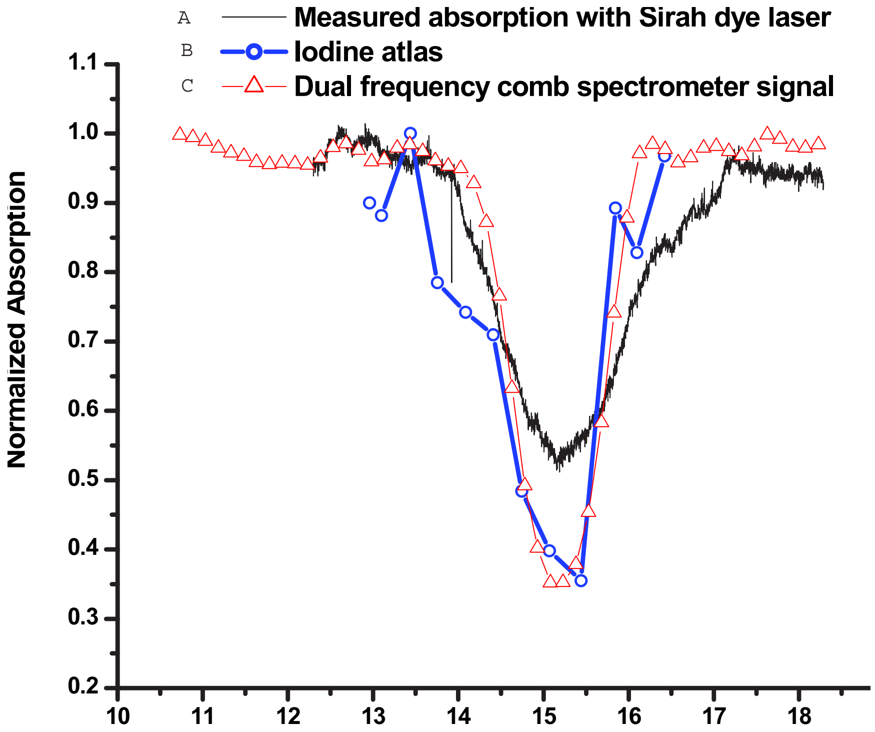

Figure 7. A: Transmission plot of the Sirah Laser beam as it is scanned over the iodine absorption feature at 16694.5 cm^{-1}. B: The plot of the transmission line at 16694.5 cm^{-1} from the iodine atlas. C: The transmission plot of a single cross beat from the dual etalon spectrometer as the Sirah dye laser is step scanned across the iodine absorption feature.

5 Future Directions

5.1 Broadband Spectrometer, Multi-heterodyne Design

The spectrometer described above is capable of providing high-resolution (20 MHz resolution) spectra over several GHz at a single time. However to be truly useful it needs to be able to provide high-resolution spectra over many thousands of GHz simultaneously. This is possible by making a simple change to the spectrometer demonstrated here. The change is to replace the short (1.5GHz) confocal etalon in the read out arm of the spectrometer with a longer etalon that has nearly the same free spectral range as the confocal etalon in the absorption arm of the spectrometer. By making the free spectral range only slightly different, say 100KHz different in frequency then the cross beat pattern between the output of the two etalons will be a series of cross beat frequencies separated by 100KHz. The number of such separated frequencies will be equal to the number of comb teeth coming from the two etalons. To obtain a 100KHz separation between the two etalons requires that the etalons approximately 10 microns different in length. Figure 8 is a schematic of the expected frequencies originating from the two etalons and the corresponding cross beat frequencies that will be observed when the output of the two etalons are combined on a fast photodiode. Note that the series of cross beat frequencies separated by (100KHz in this example) are repeated every 150 MHz. This is because there are cross beats not only between the nearest frequencies but between all frequencies that are present in each beam. In this manner one can read out the absorption spectrum over the entire bandwidth of the laser and the entire spectrum resides between 150 and 300 MHz. This does not require especially fast detector or electronics and moves the spectrum away for sources of noise caused by vibrations in the laboratory.

5.2 Time Resolved Fourier Transform Spectroscopy

As we have demonstrated above we can measure the absorption spectra with about 20 MHz frequency resolution by observing the cross beat interference pattern for about 50 nanoseconds. Therefore if we had a light source that was on for an extended period of time, say 100 microseconds, we could record the interferogram continuously for the 100 microseconds and then after the fact Fourier Transform each 50 nanosecond piece of the interferogram individually. In this way we would have a “movie” of the absorption with 50 nanosecond resolution over the entire 100 microseconds. Each individual frame of the movie would contain the entire absorption spectrum with approximately 200 MHz frequency resolution. The entire time resolves spectrum would be recorded in 100 microseconds. To obtain equivalent spectral resolution with a traditional Fourier transform spectrometer utilizing a moving mirror would require a mirror travel of fifty feet. In order to obtain the time resolution one typically performs a step scan of the mirror. That is the time changing absorption must be a reproducible event and the event, like a photolysis of a molecule to produce a radical whose changing absorption spectra one wants to record. In this case a photolysis pulse would cause the radical to be formed and the interferogram would be recorded at a set mirror position. Then the mirror moved and the photolysis repeated. This would be done until the desired resolution is obtained. This can take many hours. The comparable information

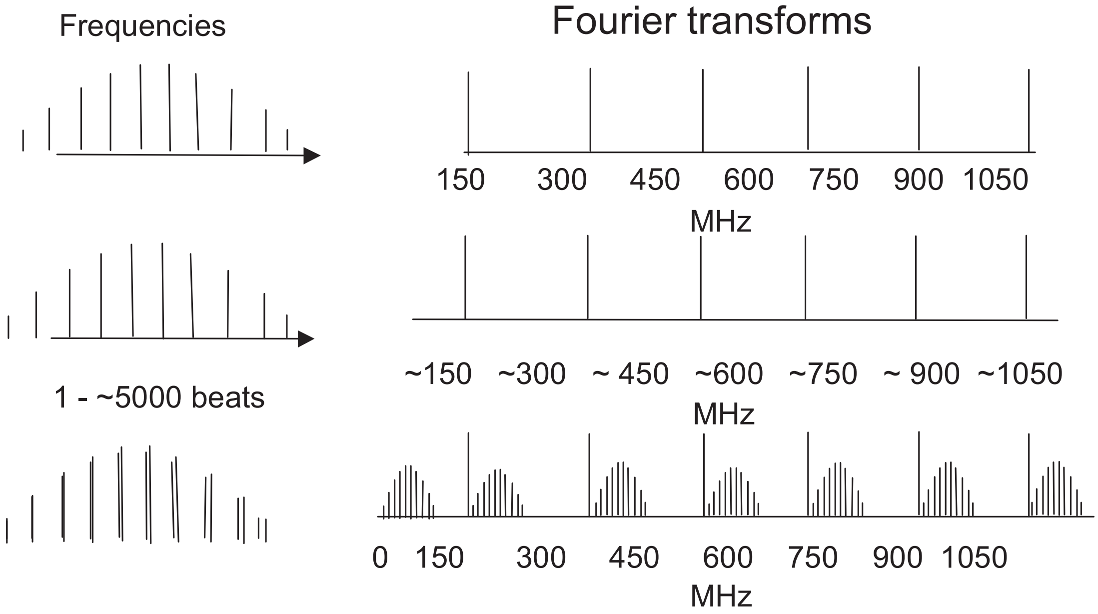

Figure 8. Schematic of the frequencies present in both the absorption arm of the spectrometer (upper panel) and the read out arm of the spectrometer (middle panel) and in the combined outputs of the two etalons. New beat frequencies are obtained in the combined signals (Lower Panel) and these new frequencies contain the entire spectrum of the parent signals from the absorption arm but shifted to lower frequency.

can be obtained with the techniques outlined here following a single photolysis pulse. If better signal to noise is needed then several interferograms from successive photolysis pulses can be averaged together. The technique outlined here is six or seven orders of magnitude faster than traditional time-resolved Fourier transform spectroscopy and over an order of magnitude higher resolution than traditional Fourier Transform spectroscopies (with bright laser sources and cavity ring down quality mirrors one can obtain 100 KHz resolution which is four order of magnitude better resolution than traditional FT spectrometers).

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