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ABSTRACT

Mid infrared frequency combs allow for high resolution absorption spectroscopy of molecular species, which have strong signatures in this spectral region. Dual comb spectroscopy can provide broadband and high-resolution capability, but requires two fully stabilized frequency combs which adds complexity to the system.

Previous work has demonstrated that frequency combs coupled with a high resolution spectrometer, consisting of a virtually imaged phased array (VIPA) along with a grating, can perform time-resolved, broadband and high-resolution absorption spectroscopy with a single frequency comb. The VIPA spectrometer disperses the spectrum in two dimensions and images it onto a focal plane detector array. If the comb teeth can be resolved, the VIPA is easily calibrated and provides comb-tooth resolved resolution and accuracy. However, in previous work, the repetition rate of the laser sources used was too low to be resolved directly, and additional passive “filter cavities” had to be employed to increase the effective repetition rate of the frequency comb. In this work we use a fully stabilized mid infrared frequency comb based on a 1.6 GHz repetition rate modelocked vertical external cavity surface emitting laser (VECSEL) and difference frequency generation to produce an offset free comb in the 3-4 micron wavelength range. The source is directly coupled to the VIPA spectrometer to provide comb-tooth resolved absorption spectroscopy. We discuss the system’s performance in gas absorption spectroscopy and its time resolving capabilities, which are limited only by the speed of the detector system.

Keywords: VECSEL, frequency comb, spectroscopy, mid-infrared, methane, VIPA, difference frequency generation, GHz

1. INTRODUCTION

Optical frequency combs provide the capability for high resolution, time resolved spectroscopy by providing a series of narrow linewidth teeth over a broad spectral bandwidth. However, detecting these comb teeth, which are often spaced by only hundreds of MHz, is a nontrivial task. One of the primary techniques is dual comb spectroscopy, which utilizes two nearly identical frequency combs to perform Fourier transform spectroscopy without a need for a moving delay stage. Recently, high resolution dual comb spectroscopy has been performed in the mid-IR.\(^1\) However, this technique typically requires two similar frequency comb systems with a slight detuning in repetition rate, which adds complexity. Single comb spectroscopy techniques relying on dispersive spectrometers, on the other hand, typically do not have sufficient resolution to natively resolve individual comb teeth without the addition of filtering cavities, which also add complexity and reduce overall power efficiency. The highest resolution spectrometers often rely on a highly dispersive element such as a virtually imaged phased array (VIPA)\(^2\) used in conjunction with a grating in the orthogonal direction to cross disperse the spectrum so that degeneracy of the VIPA orders is lifted. These have been demonstrated in the visible, near-IR, and mid-IR.\(^3\)–\(^5\) Cross dispersal spectrometers like the VIPA and related immersion grating techniques\(^6\) typically

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have spectral resolution that is around 1 GHz. This resolution is not sufficient to resolve the commonly used
fiber based frequency combs, which typically have repetition rates from 50-250 MHz. As a result, most previous
demonstrations of comb resolved spectroscopy with VIPA spectrometers have relied on filter cavities to resolve
individual comb teeth, although unfiltered combs have been resolved in the near-IR for use as a calibration
source for upconverted mid-IR combs. Time resolved measurements with these systems can allow for transient
reaction products to be detected.

The mid-infrared spectral region spanning from 2-20 microns is of special interest for frequency comb spec-
troscopy of gas species because they typically have stronger absorption in this region compared to the near-IR. In
this work, we present a system for comb resolved spectroscopy in the mid-infrared that utilizes a single frequency
comb with 1.6 GHz repetition rate based on a modelocked VECSEL as described in ref. This repetition rate is
high enough to be natively resolved with a VIPA spectrometer without the use of filter cavities, thus preserving
the full output power of the comb. This frequency comb is fully stabilized and operates in the 3-3.5 micron
region, allowing it to be used for gas spectroscopy measurements. Since the comb teeth can be resolved, the
resolution limit of the system is defined by the frequency comb itself, rather than the resolution of the dispersive
spectrometer. Methane (CH₄) features a number of strong vibrational absorption bands in the 3 micron (3333
cm⁻¹) region, making it an ideal candidate for characterization of this system.

2. EXPERIMENTAL SETUP

The system consists of a mid infrared frequency comb based on difference frequency generation operating from
3.0-3.5 microns that has been previously introduced and characterized. The comb utilizes a VECSEL that is
passively modelocked with a semiconductor saturable absorber mirror with a repetition rate of approximately 1.6
GHz and an initial output wavelength of 1030 nm. Difference frequency generation produces a comb in the mid
infrared with zero offset frequency, allowing for full stabilization of the comb with a simple repetition rate lock.
The frequency of each comb tooth can therefore be represented by $f_{comb} = N \times f_{rep}$, with $N$ a positive integer.
This direct knowledge of the frequencies of the comb teeth allows them to be used to calibrate spectrometer mea-
surements. However, comb teeth with <1 GHz spacing cannot be easily resolved with traditional spectrographic
techniques. Here, a system consisting of a VIPA tilted etalon and cross dispersing grating (figure 1) are used to
image the comb teeth in two dimensions onto an InSb cooled camera sensor (FLIR A6750SC). The VIPA etalon
is a 800 micron thick, fabricated from silicon. The input face has a small AR coated region for input coupling
of the MIR signal into the etalon, while the remaining portion of the input face has a high reflectivity coating
for the 3-3.5 micron wavelength range. The output face has a 98% reflective coating to produce the high finesse
necessary for resolving the comb teeth. The VIPA etalon is tilted at a 15 degree angle, and the input beam is
focused to a line with a 50 mm focal length cylindrical lens. The cylindrical lens is radially aligned such that the
focused line is coupled through the AR coated entrance slit on the etalon, while the beam remains collimated
in the other plane. Subsequent reflections off of the two faces of the tilted etalon produce virtual sources in the
VIPA plane. These sources interfere to produce high angular dispersion, but with diffraction orders that overlap
every free spectral range of the VIPA (approximately 54 GHz). The spectral resolution of this VIPA etalon was
experimentally determined to be 600 MHz, which is sufficient to resolve the 1.6 GHz frequency comb used here.
In order to spread out the overlapping orders, a 450 groove/mm grating designed for 3.1 μm was used in a
reflection configuration to create angular dispersion in the plane orthogonal to the VIPA dispersion. The net
effect of these two dispersive elements is a beam that is roughly collimated for a given narrow spectral bandwidth,
but with differing angles in 2 dimensions depending on wavelength. In order to turn this into spectral data with
a spatial dependence, the output of the grating is focused onto the InSb sensor directly using a combination of
a 10 inch reflected focal length 30 degree off axis parabolic mirror, and a 150 mm CaF₂ lens AR coated for the
2-5 μm region. The choice and spacing of the optics was chosen to maximize the optical bandwidth imaged on the
sensor while still allowing for resolution of individual comb teeth.

Figure 2a shows a typical VIPA image with resolved comb teeth. The InSb sensor has a 15 μm pixel pitch,
a 640 x 512 resolution, and a maximum frame rate of 125 Hz for a full frame image. The sensor was cooled to
77K with an integrated chiller. A two point calibration was performed prior to measurements to account for
nonuniformity in the detector, and a background image with the frequency comb blocked was taken to subtract
from each data set. The camera has the capability of frame rates exceeding 4 KHz for smaller window sizes,
allowing for very fast spectroscopic measurements to be performed albeit at a significantly reduced spectral bandwidth. In order for the adjacent VIPA columns to be stitched together into a contiguous set of data, the image on the sensor must contain at least 1 full VIPA FSR in the vertical direction. The 1.6 GHz comb, this corresponds to typically 33-34 visible dots per column, depending on the line. Figure 2b shows a sample with methane absorption present with repeating absorption features visible at the top and bottom of the image. These absorption measurements were conducted by passing the frequency comb through a 13 cm long gas cell with AR coated ZnSe windows. A bandpass filter with 3.46 μm center wavelength and 140 nm bandwidth was used to reduce the 500 nm bandwidth of the MIR comb for easier initial calibration of the acquired spectra. In the vertical direction, the data continues after an interval corresponding to the FSR of the VIPA. This data covers the same spectral range as the adjacent column in the original VIPA order. Therefore, to turn these 2D images into a continuous spectrum, adjacent lines must be concatenated every 54GHz. This also means that an ideal detector for this system would have enough vertical extent to fit one complete unique line in the vertical direction, and as much horizontal extent as possible in order to simultaneously measure the widest optical bandwidth possible. However, because our system utilizes an off the shelf InSb camera, it imposes limitations on the maximum spectral bandwidth that can be accessed simultaneously. Furthermore, the camera sensor is recessed approximately 19 mm behind a annular coldshield that produces the circular windowing seen in the VIPA images. This coldshield does not function as a field stop for the higher F number lenses designed to be used with this system, but for our two element setup here it is the primary limiter of the field of view of the image, and therefore the spectral bandwidth. A wider span can be accessed by changing the angle of the diffraction angle, but this requires multiple measurements and therefore doesn’t increase the bandwidth for time resolved measurements.

The repetition rate of the frequency comb was locked using a combination of a phase detector (Vescent D2-135) and external frequency generator referenced to a rubidium standard that feeds an error signal to an external servo controller (Vescent D2-125). A piezo-electric transducer mounted to a cavity mirror is used to control the repetition length by stabilizing the total cavity length. Locking the repetition rate in this manner yields a MIR
Figure 2. (a) Raw image from VIPA spectrometer with no methane present showing resolvable comb modes (b) Raw image with 300 torr pressure of 10% methane, 90% argon gas mixture equalized to 1 atmosphere, for an approximate concentration of 4% methane. The region inside the red lines correspond to unique data within 1 FSR of the VIPA etalon.
comb that is fully stabilized to within a few MHz.\textsuperscript{9}

Absorption measurements were performed on a gas mix made up of 10\% methane and 90\% argon. For a spectral measurement, the chamber is first pumped down and a reference VIPA image is taken with a 300 microsecond integration time. 1000 frames are averaged together with 125 Hz framerate to form one image. This image serves as both an amplitude reference and a frequency calibration for the VIPA system, since all comb teeth within the spectral window determined by the sensor will be visible and their frequency spacing is known to a high degree of accuracy. The chamber is then filled with the methane gas mix and a second image is taken.

3. DATA PROCESSING AND RESULTS

After each image set is averaged together to produce the reference and gas sample images, the background image is subtracted from each. Because the frequency of each comb tooth is different, each column of dots in the image will necessarily have a slight angle even when the system is completely aligned in plane with the camera due to the grating dispersion. Typically, there is an additional tilt that is caused by slight misalignment of the camera sensor relative to the other elements of the optical system. To correct for these factors and enable for proper detection of the columns, the image is first rotated by a small angle if necessary. A gaussian filter with standard deviation of .7 pixels is applied to the image to reduce noise fluctuations from pixel to pixel. A peak detection algorithm detects the locations and amplitudes of all of the peaks in the image, which correspond to resolved comb teeth. These peaks are then grouped into columns and sorted in order of increasing optical frequency based on their location in the image. The gas sample image is also rotated and noise filtered. The peak locations determined from the reference image are accessed in the sample image, and the amplitude at each location is recorded. For these results the pixel value at the referenced location (after filtering) was sufficient for good spectra, but in other situations a more complex algorithm for obtaining the peak amplitude, such as local averaging or curve fitting could be employed. These amplitudes from the sample image are divided by the
amplitudes from the reference image to yield a transmission value. By repeating this process for each frame in a video, time resolved data can be acquired.

While the frequency comb provides an absolute calibration for the frequency spacing between data points, the start and endpoints of the data must be calibrated. Previous VIPA demonstrations have used tunable single frequency lasers, but here the methane absorption lines were sufficient as a reference for calibration. Although the FSR of the VIPA is known to within 1.6 GHz, at each jump to an adjacent column the correct dot must be chosen as the next dot in the data series. Because of potentially known rotation angle of the system and variation and curvature of the lines over the span of one image, simply determining linear start and end bounds for a single VIPA FSR across the whole image is not feasible. Instead, this is accomplished by comparing the raw data to the theoretical methane absorption from the HITRAN database, and for each column choosing either the dot above or below the vertical location of the starting dot from the previous column. Through this method, the absolute optical frequency of each dot in the images is determined because of the fully stabilized nature of the comb. The fact that the comb teeth are resolved at our wavelength of interest allows for this relatively simple and accurate calibration, compared to more complex calibrations that must be performed for spectroscopy with lower repetition rate combs that cannot be resolved by the VIPA. This also removes the instrument lineshape of the VIPA system, and allows for measurements with resolution limited by the comb linewidth by scanning of the comb teeth.

Figure 3 shows a transmission measurement taken by the system with an initial gas pressure of 300 torr. The chamber was then equalized to 1 atmosphere to pressure broaden the lines and give an effective methane concentration of 4%. We can see good agreement between the measurement and the theoretical transmission, even with no scaling of the data. The main deviation from the expected transmission occurs for the most strongly absorbing peaks, where the measured data saturates at around 13% transmission in the raw data. This is possibly due to stray light from adjacent comb teeth and VIPA columns bleeding into the sampled region slightly, thereby bringing up the detection floor. Future optimizations of the optical system or utilization of fitting algorithms could likely overcome this limitation. However, we can see that the frequency accuracy of the detected peaks has strong agreement when calibrating the frequency axis using the known frequency for the repetition rate, which was locked for the duration of the measurement.

SUMMARY

In this work, we demonstrate a system for mid-IR gas spectroscopy that utilizes a single GHz repetition rate frequency comb and a high resolution spectrometer based on a virtually imaged phased array (VIPA) and InSb camera. Preliminary measurements agree well with the HITRAN predictions, and resolution of the comb teeth results in frequency accuracy and precision that is theoretically limited by the comb stability rather than the measuring apparatus. The offset free nature of the comb source allows for these measurements to be performed with only a relatively simple and robust repetition rate lock, and the video capabilities of the camera sensor show promise for time resolved measurements.

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