High Resolution Frequency Measurements of Far-Infrared Laser Lines

Elizabeth J. Ehasz, Thomas M. Goyette, Robert H. Giles and William E. Nixon

Abstract—The frequency of four previously reported far-infrared laser lines have been measured to an accuracy of 100 kHz. These laser lines were measured using a heterodyne system which allowed for more accurate measurement. The four far-infrared laser lines which originated from Formic Acid (HCOOH), O-Deutero-Formic Acid (HCOOD), C-Deutero-Formic Acid (DCOOH), and Methyl Chloride (CH_3Cl) were optically pumped by an ultra-stable, grating-tunable, CO_2 laser. Previous measurements had been conducted using Fabry-Perot cavities not typically known for their high accuracy. The difference between the frequencies measured here and the listed frequencies for these laser lines ranged from 59 MHz to 3.9 GHz.

Index Terms—FIR Laser, Gas Laser, Molecular Laser, Submillimeter-Wave Laser, Terahertz Source

I. INTRODUCTION

ACCURATE frequency measurements of far-infrared (FIR) laser lines are necessary for numerous applications. Radio astronomy, spectroscopy, imaging, and other systems using heterodyne detection rely directly on the accuracy of the laser frequency which is often used as a reference for other measurements.

In radio astronomy, a telescope will collect any number of frequencies from a particular region in space. Traditionally, the goal is to look for a specific transition in a particular molecule. In order to locate this transition, the frequencies gathered from the radio telescope are mixed with a known frequency source. Intermediate frequencies (IF) are consequently created and measured. A well-known reference frequency will allow for not only the detection of the molecule, but also its environmental conditions and relative motion. Therefore, accurate knowledge of the known source's frequency is very important. In previous work, it was determined that the reported frequency of Trideuteriomethanol (CD_3OH) 1456.7 GHz line used for radio astronomy differed from the measured frequency by approximately 2.7 GHz [1].

While some far-infrared laser lines have previously been reported to a high accuracy, there are still a large number which have not been measured to the same accuracy. Here, the frequency of four FIR laser lines originating in Formic Acid

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(HCOOH), O-Deutero-Formic Acid (HCOOD), C-Deutero-Formic Acid (DCOOH), and Methyl Chloride (CH_3Cl), which were previously measured only by coarse methods [2]-[3], are reported with higher accuracy (1 part in 10^7). These lines were measured in order to conduct spectroscopic research which required high precision knowledge of the laser frequencies.
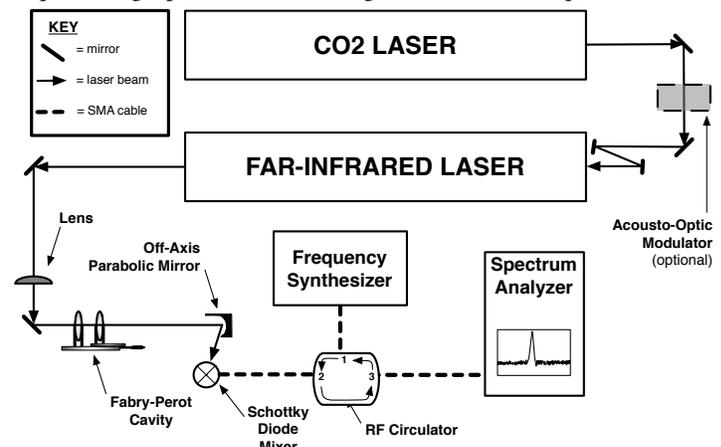

Fig. 1: A diagram of the experimental set-up used for these measurements.

II. DESCRIPTION OF EXPERIMENT

Figure 1 shows the layout for this experiment. An ultra-stable, grating-tunable, CO_2 laser with an average line width of 130 kHz was used to optically pump the sample gas in the FIR laser. The FIR laser consists of a water-cooled quartz tube with an off-axis input coupling hole and a flat output coupler. The output coupler's design varies between the more traditional hole coupler and a uniform output coupler that uses metal strips optimized for specific wavelengths.

The ultra-stable CO_2 laser consists of an 8-ft.-long vacuum section containing a high voltage DC discharge. The CO_2 laser was designed to be ultra stable, with a drift of less than ±0.5MHz for days at a time [4]. For the majority of the measurements, the CO_2 laser beam was sent directly into the FIR laser through focusing optics. The output power for the CO_2 laser was between 120W and 150W depending on the pump line chosen.

An optional acousto-optic modulator (AOM) was employed to pump the Methyl Chloride FIR laser line in an effort to stabilize the far-infrared laser's signal. The AOM is a water-cooled single germanium crystal, activated by a piezoelectric transducer (PZT). Acting as both a Bragg scatterer and a frequency modulator, the AOM has the ability to diffract an incident beam as well as shift the frequency of the incident beam by the amount of the acoustic frequency [5]. Stability

of the far-infrared laser's signal was increased by reducing the offset between the CO_2 laser's center frequency and the center absorption frequency of the Methyl Chloride in the far-infrared laser. This was accomplished by sending the CO_2 laser beam through the AOM and shifting the frequency by 40 MHz.

The FIR laser lines measured were produced using a highly stable, far-infrared molecular-gas laser designed and built at the University of Massachusetts Lowell [6]. The FIR laser consists of an 8 ft. long quartz tube with flat mirrors forming the laser cavity. The CO_2 laser entered the cavity through a 2 mm diameter off-axis hole in the input coupler. The output coupler has an on-axis 2 mm diameter hole through which the FIR laser beam was emitted. The diameter of the input and output coupler is 2 inches. The walls of the FIR cell were designed as dielectric waveguides to overcome diffraction losses. FIR laser tubes, acting as dielectric waveguides, have been described previously [7] - [8]. Both the tube and the optics were water-cooled to approximately $13°C$ to increase stability through reduced thermal expansion drift. The FIR laser operated with the sample gas sealed off inside the vacuum cell at the optimum pressure, which ranged from 100 mTorr to 350 mTorr, depending on the sample gas.

A preliminary measurement of the laser wavelength was made by using the Fabry-Perot (FP) cavity shown in Figure 1. This process was necessary to verify that the wavelength detected was the correct wavelength, and not a competing FIR laser transition which lases with the same CO_2 pump line. O-Deutero Formic Acid can produce two different frequencies when pumped by the 10R22 CO_2 pump line.

The FIR laser beam was focused onto a corner cube mounted, whisker contacted, Schottky diode. The Schottky diode was used as both the heterodyne detector and multiplier in this system. The output of a frequency synthesizer was applied to the diode using a circulator. The beat frequency between the nearest harmonic of the synthesizer and the FIR laser was measured using a spectrum analyzer.

Since the diode also acted as a frequency mixer, the frequency comb from the synthesizer was combined at the diode with the FIR laser frequency (f_l) to produce an intermediate frequency [9] - [10],

$$f_{IF} = mf_l \pm nf_o, \tag{1}$$

where, for the purposes of this experiment, $m = 1$, $n =$ an integer harmonic number, and $f_o =$ the synthesizer frequency.

Before measuring the intermediate frequency, calculations were carried out in order to have an estimate of what the intermediate frequency would be. This calculation allowed for a starting location from which the search for the intermediate frequency would commence.

Predicted frequencies of the FIR laser transitions were obtained from the CRC Handbook of Laser Lines [11]. Using equation (1), the synthesizer's frequency was chosen so that the intermediate frequency would be between 8 and 15 GHz. This was accomplished by choosing a frequency which, when multiplied by an integer, would yield a frequency within 8 to 15 GHz of the predicted FIR laser transition.

The intermediate frequency was sent to the spectrum analyzer through the circulator for measurement. Since the synthesizer's frequency was well known (better than 1 Hz) the laser frequency was accurately determined by measuring the intermediate frequency and solving for f_l in equation (1).

The harmonic number, n, was determined by adjusting the frequency synthesizer's output, f_o, by a small amount, $\triangle f_o$, which corresponded to a small change in the intermediate frequency on the order of

$$f'_{IF} = f_{IF} \pm n\triangle f_o. \tag{2}$$

From this adjustment, the harmonic number n, and whether the FIR laser frequency was above or below the synthesizer's harmonically multiplied frequency were obtained.

The frequency of the previously reported FIR laser lines in this study were typically known to only ± 1 GHz. Since the spectrum analyzer is capable of measurement accuracies of better than 100 kHz it was necessary to set it to a very slow frequency sweep with high resolution bandwidth in the predicted frequency range of the intermediate frequency.

The resolution of the spectrum analyzer was adjusted to 10 kHz in order to make an accurate measurement of the intermediate frequency. The resolution of the frequency measurement was directly related to both the resolution of the spectrum analyzer's display and the stability and width of the FIR laser transition, which may fluctuate by ± 30 kHz due to mechanical noise.

III. RESULTS

The results of the experiment are listed in Table I. For reference, information from the CRC Handbook of Laser Lines [11] is shown. It was from these listings that the initial predictions of the intermediate frequencies were made. The Harmonic Number refers to the harmonic, n, by which the synthesizer's frequency, f_o, was multiplied.

In order to measure the Methyl Chloride line, the CO_2 laser beam was propagated through an acousto-optic modulator, shown in Figure 1, in order to shift the CO_2 laser center frequency by approximately 40 MHz closer to the ideal center pump frequency of the Methyl Chloride absorption line in the FIR laser.

The Formic Acid line does have a competitor [11] (398.1 μm; 753 GHz) with the same CO_2 laser pump line, 9R14. However, prior to measuring the laser line with the spectrum analyzer, the Fabry-Perot cavity was used to verify that the wavelength was relatively close to the predicted wavelength. A uniform output coupler was used to lase this FIR line. This method helped suppress the competing line, as the reflectivity was not suitable for the competitor line to lase.

The measurements for this experiment have an uncertainty of approximately 100 kHz. Previous measurements reported in [11] were typically measured using Fabry-Perot interferometers [2] - [3]. In one case [3] the FIR laser itself was used as a Fabry-Perot interferometer. Typical uncertainties using Fabry-Perot cavity measurements are about $\pm 0.1\%$ due to the constraints of measuring changes in cavity lengths, implying uncertainties of ± 1 GHz in the frequencies reported in [11].

Far-infrared Laser Frequency Measurements

Gas	Listed Wavelength [1] (μm)	Listed Frequency [2] (GHz)	CO_2 Pump Line	Harmonic Number n	Measured Frequency (GHz)	Measured Wavelength [3] (μm)	Measured *minus* Listed (GHz)
Formic Acid (HCOOH)	336.3	891.	9 R 14	59	894.974227(100)	334.973286(100)	+ 3.974
O-Deutero Formic Acid (HCOOD)	351.9	852.	10 R 22	60	849.829463(100)	352.767785(100)	- 2.171
C-Deutero Formic Acid (DCOOH)	341.8	877.	10 R 16	54	878.882035(100)	341.106594(100)	+ 1.882
Methyl Chloride (CH$_3$Cl)	334.0	897.7	9 P 42	59	897.759382(100)[a] 897.759948(100)[b]	333.934085(100) 333.933874(100)	+ 0.059

[1] - wavelength as reported by the CRC Handbook of Laser Lines [11]

[2] - frequency as reported by the CRC Handbook of Laser Lines [11]

[3] - as calculated from the measured frequencies

[a] - measurement of Methyl Chloride line with acousto-optic modulator

[b] - measurement of Methyl Chloride line without acousto-optic modulator

TABLE I: RESULTS OF EXPERIMENT TABULATED

The method used in this experiment was more accurate since the FIR line was measured via a well known intermediate frequency. Uncertainties described are due to the mechanical vibrations on the FIR laser which manifest themselves as a frequency jitter of ±30 kHz.

Table I shows the comparison of the measured frequency, using the methods described, and the listed frequencies from [11]. The last column of Table I represents the Measured Frequencies minus the Listed Frequencies. These values range from quite good (59 MHz for the Methyl Chloride line) to relatively large (3.9 GHz for the Formic Acid line). On average, this variation is consistent with the typical FP uncertainty of about 0.1%, although the difference of 3.9 GHz is considered somewhat of a surprise since it is a proportional difference of 0.4%.

IV. DISCUSSION

The O-Deutero Formic line was an interesting case. This line was run with a uniform output coupler, but originally the coupler was in the wrong orientation to run this line. The output coupler in question would have had a reflectivity of 95% for a parallel polarization line and 19.5% for a normally polarized line. 19.5% is normally insufficient to produce lasing. However, the line was able to still run, and with extraordinary power considering the circumstances. The signal detected on the diode was 0.8V$_{pp}$, equivalent to 1.1mW of power, with the output coupler in the wrong orientation.

A wire grid polarizer was used to determine that the beam was in fact vertically polarized. It was also noted during the experiment the O-Deutero Formic line had a moderate offset which made the far-infrared laser line slightly unstable. While the offset was not measured, it was clear that the offset was less than the 40 MHz offset of the Methyl Chloride line.

The output coupler's orientation was then corrected, and measurements were repeated. The signal detected on the diode was 1.2V$_{pp}$ which is equivalent to 1.7mW of power. While the line was more powerful with the output coupler in the correct orientation, the frequency measurements were the same with both orientations of the output coupler.

V. CONCLUSION

Frequency measurements of far-infrared laser lines were made with an accuracy of 1 part in 10^7. The measurements were acquired by mixing the far-infrared laser signal on a Schottky diode with signal from a frequency synthesizer. From these two frequencies, an intermediate frequency was produced. It was this intermediate frequency that was ultimately detected and the far-infrared frequency thus calculated with high accuracy. In all, four far-infrared laser lines were detected in this manner and measured. A frequency accuracy of ±100 kHz was achieved. Measured frequencies differ from previously reported measurements by as much as 3.9 GHz.

ACKNOWLEDGMENT

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