The Generation and Frequency Measurement of Short-Wavelength Far-Infrared Laser Emissions

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ABSTRACT

A significantly improved three-laser heterodyne system has been assembled to generate and measure short-wavelength far-infrared (FIR) laser emissions. Over the past several years, this system has been used to discover fifty-five FIR laser emissions, ranging in wavelength from 26.3 to 185.0 μm. These emissions were generated by optically pumping a FIR cavity with a continuous-wave carbon dioxide laser in a X-V geometry when using either hydrazine or a methanol isotopologue as the FIR laser medium. Although heterodyne techniques can be used to measure the frequencies of these FIR laser emissions with fractional uncertainties of $\pm 2 \times 10^{-7}$, shortcomings in the previous system limited its effectiveness. Improvements made to the three-laser heterodyne system have resulted in an increase in the spectral range used to search for the beat between the known and unknown laser frequencies (an increase of up to 25 GHz) as well as an increase in the system's sensitivity (by up to a factor of 30), all without requiring the use of an additional microwave frequency source. With this improved system, the FIR laser frequencies for the recently discovered 53.9 and 90.0 μm laser emissions generated by optically pumped CH_3OD have been measured.

INTRODUCTION

The discovery of the optically pumped molecular laser (OPML) by Chang and Bridges in 1970 [1] constituted the decisive step toward developing a coherent radiation source that could provide useful coverage of the far-infrared (FIR) spectral region. Since then, over 5,000 OPML emissions have been discovered, ranging in wavelength from approximately 10 to 3030 μm (some useful reviews of known FIR laser emissions include Refs. [2-5]). Recently, an OPML system was developed for the generation of short-wavelength FIR laser emissions ($\lambda <$ 150 μm). With this system, fifty-five optically pumped FIR laser emissions, having wavelengths ranging from 26.3 to 185.0 μm, were discovered from hydrazine (N_2H_4) and isotopic forms of methanol (CD_3OH, $^{13}CH_3OH$, $^{13}CD_3OD$, CH_2DOH, CHD_2OH, CH_3OD and $^{13}CD_3OH$) [6-11]. This includes the discovery of the 32.8 μm line, the shortest-known OPML emission generated by CHD_2OH [9]. The motivation for these investigations was that of the known OPML emissions, less than 25% were present in the short-wavelength portion of the FIR region. Of these lines, less than 30% have had their frequencies measured. Consequently, a three-laser heterodyne system was assembled, resulting in the measurement of thirty-three FIR laser frequencies [12-15]. However, the frequencies of several short-wavelength laser emissions, including the recently discovered 53.9 and 90.0 μm laser lines from optically pumped CH_3OD, were unable to be measured due to limitations with the system [12]. Therefore, to improve on the effectiveness of using OPML systems in this portion of the electromagnetic region, the performance of the existing three-laser heterodyne system was re-evaluated and its ability to measure the frequencies of short-wavelength FIR laser emissions was improved. In this paper, an overview of the three-laser heterodyne system, including the modifications made to improve the system's spectral range and sensitivity, will be discussed. Also reported are the FIR laser frequencies measured with this system for the previously mentioned CH_3OD emissions along with the offset of the carbon dioxide (CO_2) pump laser from its center frequency for each FIR laser emission.

EXPERIMENTAL DETAILS

I. Generation of FIR Laser Emissions

A three-laser heterodyne system has been assembled to generate FIR laser emissions and measure their frequencies. The heterodyne system consists of an optically pumped molecular laser and two additional CO_2 lasers, as shown in Fig. 1. FIR laser emissions are generated by optically pumping the laser medium in the FIR cavity with

a continuous-wave CO_2 laser. The CO_2 laser is 2 m long and uses a Pyrex glass tube containing equally spaced glass ribs surrounded by a water-cooled jacket. The laser uses a 133 line/millimeter grating that provides approximately 3% output coupling in zeroth order. Both the 9 and 10 μm branches exhibit lines out to 9*R*58, 9*P*60, 10*R*58 and 10*P*60, with powers up to 30 W [16-18].

The CO_2 laser radiation is focused into a 2 m long, nearly confocal FIR cavity using a X-V pumping geometry [6]. This geometry, illustrated in Fig. 1, uses three copper mirrors, 19 mm in diameter, a gold-coated copper mirror with a radius of curvature of 1 m and one of the FIR cavity mirrors. Once entering the FIR cavity, the CO_2 radiation is first reflected across the vertical plane of the cavity by a 45° mirror. At the other end, two identical 45° mirrors redirect the CO_2 beam to the bottom of the input chamber. The gold-plated copper mirror then reflects the CO_2 beam to the main FIR cavity mirror. The CO_2 beam is reflected from the FIR mirror, to the input 45° mirror, and out of the FIR system. Conversely, the FIR laser radiation is coupled horizontally out of the cavity, through a polypropylene window by means of a 45° copper mirror and focused by an off-axis parabolic mirror onto a tungsten-nickel metal-insulator-metal (MIM) point-contact diode.

Preliminary wavelength measurements of the FIR laser radiation were made by tuning the Fabry-Perot cavity with the moveable end mirror and measuring the mirror displacement of at least twenty adjacent longitudinal modes for a particular laser emission. This value obtained for the wavelength has an uncertainty of ± 0.5 μm. A set of absorbing filters, calibrated with wavelength, attenuates the CO_2 laser radiation and helps distinguish different FIR wavelengths. The relative polarizations of the FIR emissions with respect to the CO_2 laser lines were measured with either a multi-Brewster-angle polarization selector or a gold wire-grid polarizer (1000 lines per inch) [6-11].

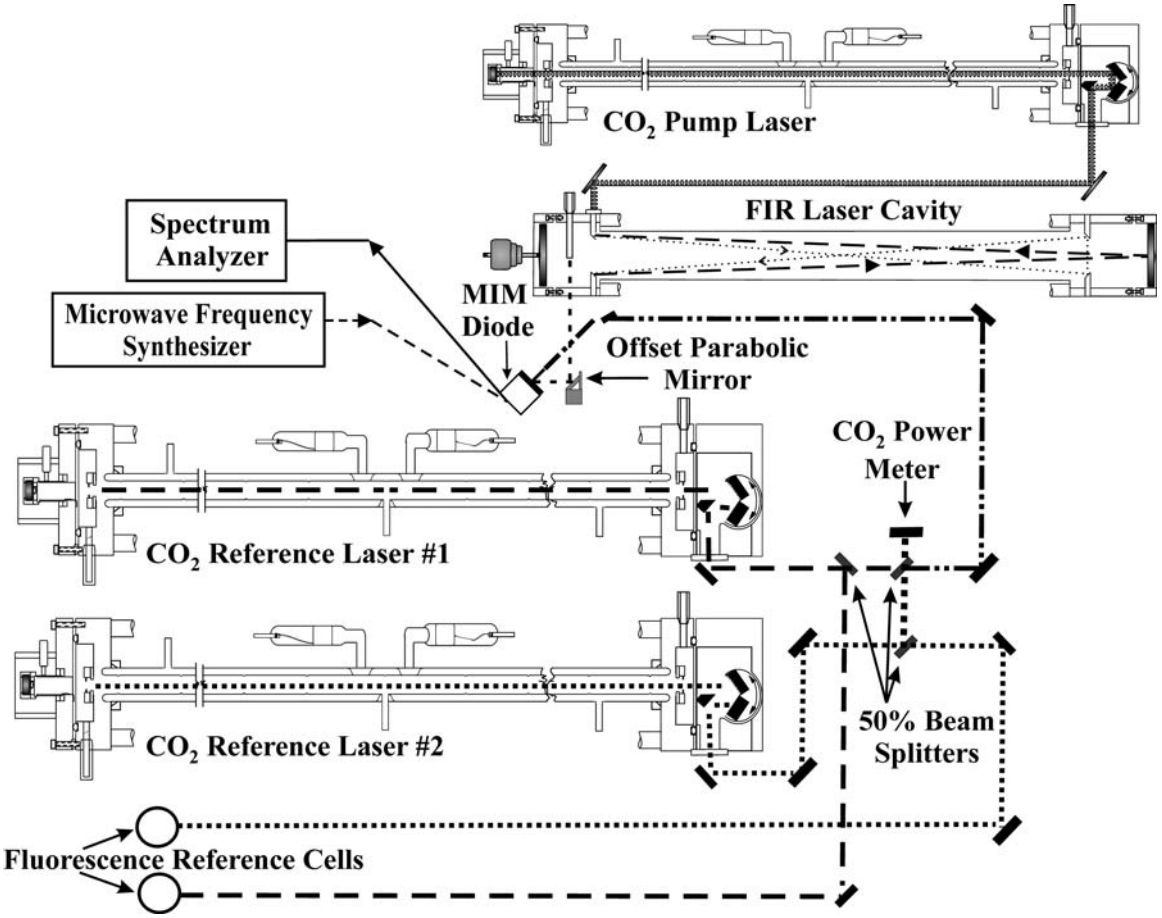

Figure 1. The three-laser heterodyne system.

II. Measurement of FIR Laser Frequencies

The FIR laser frequencies were measured using the three-laser heterodyne technique, discussed in detail in Refs. [8, 19, 20]. Two CO_2 laser frequencies were combined in the MIM diode to create a difference frequency in the FIR region. These CO_2 reference frequencies were chosen such that their difference frequency nearly equals the FIR laser frequency calculated from the wavelength measurement. A beat between this difference frequency and the unknown FIR laser emission (also focused on the MIM diode) was then amplified and observed on a spectrum analyzer. If the separation between these frequencies was greater than the range of the spectrum analyzer, a microwave source was added to the MIM diode to decrease the frequency gap. The frequency of the beat was then measured by comparison with standardized frequency markers generated by the spectrum analyzer.

This measurement determined the unknown FIR laser frequency, ν_{FIR}, through the relation

$$\nu_{FIR} = | n_1\nu_{CO(I)} - n_2\nu_{CO2(II)} | \pm m\nu_{\mu wave} \pm \nu_{beat}, \qquad \text{Eq. 1}$$

where $| n_1\nu_{CO(I)} - n_2\nu_{CO2(II)} |$ is the difference frequency synthesized by two CO_2 lasers, $\nu_{\mu wave}$ is the microwave frequency and ν_{beat} is the beat frequency. The integers, n_1, n_2 and m define the harmonics of the difference and microwave frequencies, respectively (fundamental, first harmonic, etc.). The value of m and the ± signs in Eq. 1 were determined experimentally by tuning the FIR laser cavity and slightly varying the microwave frequency to obtain a small shift in the beat, as observed on the spectrum analyzer.

The CO_2 reference lasers were frequency stabilized to their saturation dips in the 4.3 μm fluorescence signal from an external reference cell [21]. The one-sigma fractional uncertainty, $\Delta\nu/\nu$, of FIR laser frequencies measured with this technique was $\pm 2 \times 10^{-7}$. This uncertainty was derived from setting the FIR laser cavity to the center of its gain curve. To minimize this uncertainty, the FIR laser was tuned across its gain curve, the center of which was measured on the spectrum analyzer using a peak hold feature. An iris in each of the laser cavities in the three-laser heterodyne system was used to eliminate higher order modes and help shape the gain curve to a symmetric pattern. The FIR laser frequency was calculated from the average of at least fifteen measurements, several of which were recorded with varying microwave frequencies. In addition, these measurements were made with at least two different sets of CO_2 reference laser lines.

Lastly, the pump offset from the CO_2 line center (the difference between the CH_3OD absorption frequency and the center frequency of the CO_2 pump laser line) was measured, which is important for assigning FIR laser transitions. Measuring the offset requires setting the pump frequency for maximum FIR power and then mixing, in the MIM diode, some of the pump radiation with a reference laser locked to the line center of the CO_2 pump laser emission. As with the FIR frequency measurements, the MIM diode generates a beat between the two laser frequencies that is measured on the spectrum analyzer. Offset frequencies were measured within ± 10 MHz (this uncertainty was derived primarily from the reproducibility of these measurements).

III. Improvements to the Heterodyne System

The original setup for the three-laser heterodyne system used a HP8558B spectrum analyzer (having a 100 kHz to 1500 MHz spectral range), a HP8672A synthesized signal generator (operating between 2 and 18 GHz), a HP8640B signal generator (having a range from 500 kHz to 1024 MHz) and an Avantek amplifier (operating between 100 kHz and 1200 MHz). As mentioned, once FIR laser emissions were detected, their wavelengths were determined to ± 0.5 μm by traversing at least twenty adjacent longitudinal modes. For short-wavelength lines, this translates into a large frequency region in which to search for the beat. As an example, the predicted laser frequency for a 50 μm wavelength measurement would have a ± 60 GHz uncertainty, which is well beyond the range of the system. Secondly, while strong short-wavelength FIR laser emissions do exist, the majority of lines recently discovered in this portion of the FIR region have had relatively modest powers, typically below 100 μW. In addition, for wavelengths below 55 μm, the first harmonic is required to generate the CO_2 difference frequency. These weaknesses, combined with the increased reliance on the additional microwave frequency source, limited this system's ability in detecting and measuring the frequencies of short-wavelength FIR laser emissions. Therefore, our goal was to increase the spectral range and sensitivity of the heterodyne system.

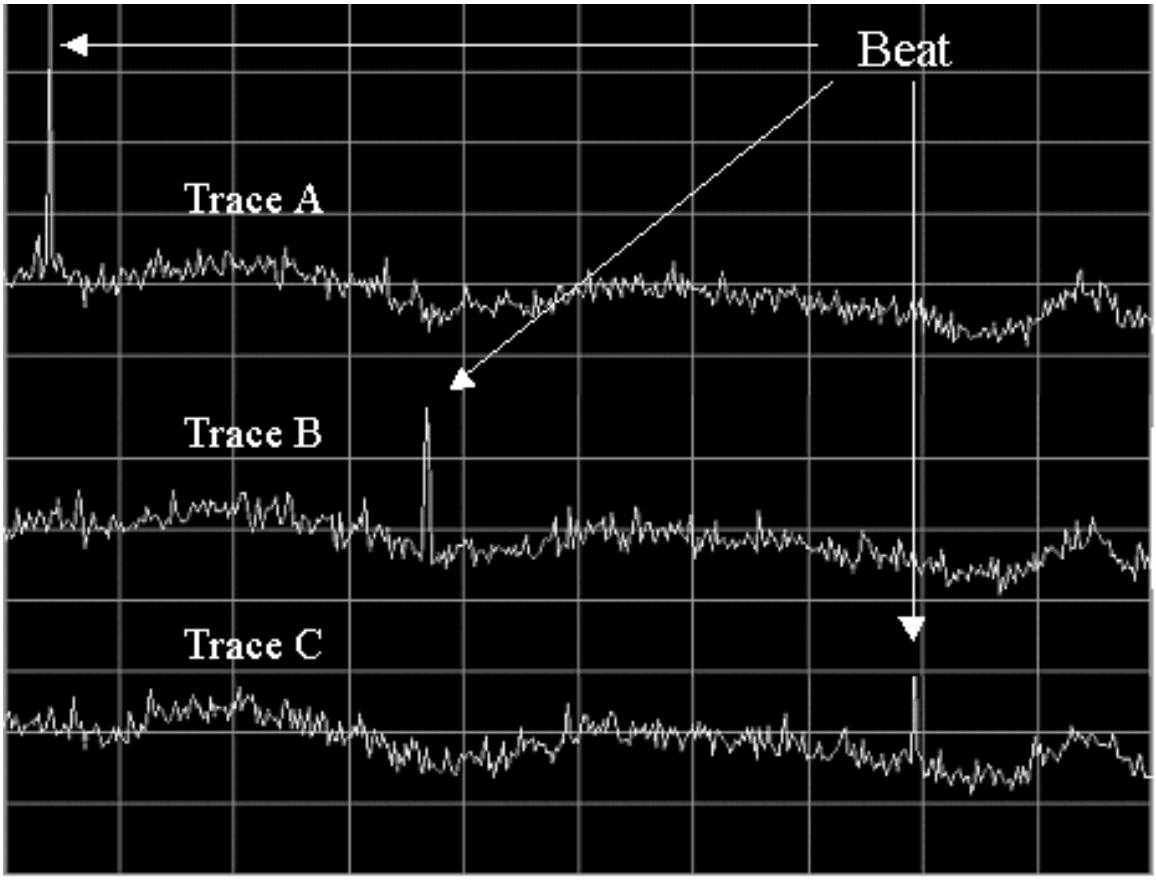

Figure 2. Trace A is for a beat frequency of + 2241 MHz (at –31 dBm, with CO_2 reference lines 9*P*10 and 10*R*4), Trace B is for a beat frequency of –10456 MHz (at –46 dBm, with CO_2 reference lines 9*P*32 and 10*P*20) and Trace C is for a beat frequency of + 21042 MHz (at –50 dBm, with CO_2 reference lines 9*P*44 and 10*P*32). The beats were observed with no additional microwave signals added and the 109.974 μm CH_3OD emission was generated using the 10*R*44 CO_2 pump line.

The improvements made to the three-laser heterodyne system include the use of a new spectrum analyzer (an Agilent E4407B having a 9 kHz to 26.5 GHz spectral range) combined with a new amplifier (Miteq AFS44 amplifier operating in a range from 0.1 to 26.5 GHz). These improvements have increased the spectral range used to search for the beat by approximately 25 GHz and have increased the system's sensitivity by up to a factor of 30. Due to the increased spectral range, the use of the additional microwave frequency source is no longer required and has become optional. This is advantageous because the additional microwave signal does not always efficiently couple into the MIM diode, resulting in an undetectable beat. The upgraded spectrum analyzer also has an internal frequency standard, thus eliminating the need for an external signal generator to accurately measure the beat frequency. The system's effectiveness in observing a beat frequency, without the use of additional microwave signals, for a short-wavelength FIR laser emission is illustrated in Fig. 2. It shows three beats between the 109.974 μm line from optically pumped CH_3OD and three different pairs of stabilized CO_2 reference laser emissions observed with beat frequencies of +2241, –10456 and +21042 MHz, respectively. Although some attenuation in the signal strength exists with increasing beat frequency, this setup represents a significant improvement over the previous system, which measured comparable beat frequencies[1] on the order of –60 to –70 dBm as opposed to the –31, – 46 and – 50 dBm signals shown in Fig. 2.

[1]Of course, due to the range of the external signal generator used in the previous system, only beat frequencies out to 1024 MHz could be measured directly on the spectrum analyzer.

RESULTS

The measured FIR laser frequencies from optically pumped CH_3OD are listed in Table I, arranged in order of the CO_2 pump line. The laser and offset frequencies are reported with their corresponding wavelength and wavenumber, calculated from the average laser frequency using 1 cm^{-1} = 29 979.2458 MHz. All FIR laser frequencies were measured under optimal operating conditions and the CH_3OD, 99% D enriched, sample was obtained from Cambridge Isotope Laboratories.

Although these FIR laser emissions were discovered with this OPML system [10], frequency measurements could not be performed due to the weak intensity of these lines [12]. For example, the power of the 90.022 μm emission was observed to be only several μW. With the improvements made to this heterodyne system, the measurements of these unknown FIR laser frequencies and CO_2 offsets were performed. Only the magnitudes of the CO_2 offsets were reported; due to their position at the edge of the CO_2 pump laser's gain curve, it was not possible to reproduce a distinctive shift in the beat (either towards higher or lower frequency) upon slight variation of the CO_2 pump laser's frequency.

Table I
Measured Laser frequencies Generated by Optically Pumped CH_3OD

CO_2 Pump	Wavelength (μm)	Frequency (MHz)	Wavenumber (cm^{-1})	Offset[a] (MHz)	Ref.
9*R*16	53.909	5 561 117.7 ± 1.2	185.4989	|33|	10
10*R*42	90.022	3 330 214.8 ± 0.8	111.0840	|30|	10

[a] Measured to within ± 10 MHz

CONCLUSIONS

In this work, an improved three-laser heterodyne system has been used to measure the frequency and offset for two recently discovered FIR laser emissions generated by CH_3OD. These frequencies were measured for the first time with the highest precision possible (to fractional uncertainties of $\pm 2 \times 10^{-7}$). With this system, the difficulties of measuring FIR laser frequencies for emissions having either low output powers and/or requiring the use of an additional microwave frequency source have been eliminated. Finally, this improved system can be used to more effectively measure the frequencies of short-wavelength laser emissions, resulting in an increase in the number of coherent FIR radiation sources for spectroscopic applications (such as laser magnetic resonance spectroscopy) and metrology (as local oscillators) in this portion of the electromagnetic spectrum.

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