FAR-INFRARED FREQUENCY MEASUREMENTS USING THE THREE-LASER HETERODYNE TECHNIQUE

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ABSTRACT

An optically pumped molecular laser system has been used to generate short wavelength laser emissions in the far-infrared (FIR) region. In this experiment, a 1.5 m long carbon dioxide (CO_2) laser was used to optically pump a 2 m long FIR cavity containing the methanol isotope CD_3OH. The FIR laser utilizes an X-V pumping geometry, recently shown to efficiently pump short wavelength emissions below 100 μm. Using the three-laser heterodyne technique, the frequencies of four distinct laser emissions, ranging between 67 μm and 105 μm, have been measured and are reported with a fractional uncertainty of $\pm$ 2 x 10^{-7}, permitting spectroscopic assignment of the laser transition.

INTRODUCTION

A significant part of our understanding of the physical world is through the application of measurement techniques. With the invention of the LASER (Light Amplification by Stimulated Emission of Radiation) in the 1960's, and developments in detector technology, the far-infrared region (FIR), ranging from approximately 10 μm to 1000 μm, has emerged as a viable area of study. Investigations of the FIR region with this coherent source of radiation were first performed with the electrical discharge laser. This type of laser is capable of producing approximately 340 emissions, including several very important lines from HCN, DCN, and H_2O vapor. In 1970, a pulsed emission from methyl fluoride (CH_3F) pumped by a CO_2 laser constituted the first optically pumped FIR laser. The optically pumped molecular laser (OPML) utilizes the superposition of infrared laser emissions with the absorption spectrum of the optical cavity's lasing medium. This technique quickly overshadowed the direct discharge laser because of the immense number of laser emissions it was capable of producing. Currently, there are over 5000 known OPML emissions in the FIR region.

Heidi and Mike graduated with B.S. degrees in physics from the University of Wisconsin-La Crosse. This research was performed during the summer of their junior year, when the worked with scientists at the National Institute of Standards and Technology in Boulder, CO, sponsored in part by NSF and WSGC/ NASA. Heidi is currently pursuing her Ph.D. in optics at the School of Optics/CREOL, University of Central Florida. Mike is currently pursuing his Ph.D. in optics at the Optical sciences Center, University of Arizona.

Following the initial observation of FIR laser emissions from methanol (CH_3OH) [1], methanol and its isotopes have produced more than 2000 FIR laser lines in the wavelength range 21.7 μm – 3030.0 μm, with CD_3OH contributing over 400 emissions alone. [2-4] The richness of FIR emissions is principally due to the excellent overlap between the CD_3-deformation region (1068.2 cm^{-1}) and the C-O vibrational band (984.4 cm^{-1}) with the 9 μm and 10 μm CO_2 laser bands, respectively. [5, 6] This overlap, combined with the torsional motion exhibited by CD_3OH, makes this molecule a prolific source of FIR laser emissions.

In this paper, we discuss the experimental setup of the OPML and three-laser heterodyne systems and report the measured frequencies of the optically pumped FIR laser emissions from CD_3OH. The coherence of the continuous-wave laser's radiation is most accurately measured using direct frequency measurements, which do not suffer from the limitations of wavelength measurements.

THE EXPERIMENT

CO_2 Laser System

There are three necessary components to any laser system: an optical cavity, a pumping mechanism, and a lasing medium. The CO_2 pump laser, shown in Fig. 1, is 1.5 m long and includes a partially ribbed cavity surrounded by a water-cooled jacket. The pumping mechanism is in the form of a 13,000 V power supply capable of providing 50 mA to each cathode. The laser medium is a flowing gas

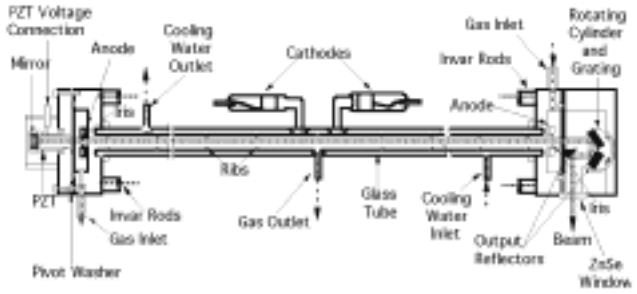

Figure 1
Top view of the CO_2 laser.

mixture of CO_2, nitrogen, and helium in the ratio 1:1.2:7.8 with CO_2 producing the laser emissions while helium and nitrogen act as buffer gases. The Pyrex glass tube has an inner diameter of 18 mm and contains five equally spaced glass ribs whose inner diameters increase from 16.5 to 17.5 mm. By introducing glass ribs into the laser cavity, many wall bounce modes are eliminated, thereby forcing an open structure mode and increasing the effective resolution of the grating. [7] The laser uses the zeroth-order output coupling from a 133 line/mm grating with 3% output coupling in zero order. A high-reflectivity gold-coated 20-m radius-of-curvature mirror, attached to a piezo-electric transducer (PZT), is used on the other end. The PZT permits the fine adjustment of the end mirror, allowing the CO_2 laser radiation to be kept tuned to its center frequency.

The CO_2 laser is capable of producing approximately 275 lines, including emissions from the 'regular', 'hot', and sequence bands. These bands indicate the vibrational levels responsible for laser action. The vibrational transitions in question are centered at 9.5 μm and 10.4 μm for the regular band, 9.3 μm and 10.8 μm for the hot band, 9.43 μm and 10.4 μm for the sequence band. [4] Within each of these vibrational transitions are two allowed possibilities for the change in the rotational quantum number, J, namely $\Delta J = J_{upper} - J_{lower} = -1$ (P-branch) or $\Delta J = +1$ (R-branch). Conventionally, a transition is designated by (for example) 10P(34), indicating the 10 μm regular band P-branch transition from the $J_{upper} = 33$ to the $J_{lower} = 34$ level. [10HP(34) would indicate a hot-band emission while 10SP(34) would indicate a sequence-band emission.] Both the 9 μm and 10 μm branches exhibit laser emission out of 9R(58), 9P(60), 10R(58) and 10P(60) with powers up to 30 W for the regular laser lines and 10 W for the hot-band and sequence-band emissions.[8]

OPML System

The experimental setup for the OPML system is partially shown in Fig. 2. The CO_2 laser radiation is focused into the FIR cavity with a 12 m radius-of-curvature gold coated concave mirror, externally mounted on the far (fixed mirror) end of the cavity at approximately 2 m from the 20 mm diameter ZnSe window. A flat gold coated mirror then reflects the CO_2 beam into the FIR cavity and to the X-V mirror system. This mirror system, shown in Fig. 2, uses three 19-mm diameter copper mirrors along with one of the FIR cavity mirrors. Each of the four copper mirrors extend about 20 mm into the FIR cavity and transmit approximately 99% of the 118.8 μm line of CH_3OH through the FIR window. (The 118.8 μm line of CH_3OH is the strongest known optically pumped FIR laser emission.) The advantage to this is that many of the longer wavelength lines are suppressed, thereby allowing some of the shorter wavelengths to emerge.

In the pumping geometry, shown in Fig. 2, a 45° mirror first reflects the beam across the vertical plane of the cavity (path 1). At the other end, two identical 45° mirrors are used to redirect the CO_2 beam to the bottom of the input chamber (paths 2 and 3 which complete the X-portion of the pumping scheme). A gold-plated copper mirror with a 1 m radius of curvature then reflects the CO_2 beam to the main FIR cavity mirror (path 4). This curvature was chosen so that the beam diameter would be slightly larger than a 40 μm Gaussian beam waist. This optimizes the overlap between the CO_2 laser beam and the short wavelength FIR emissions. Finally the CO_2 beam is reflected from the FIR mirror, to the input 45° mirror, and out of the FIR system (path 5 which completes the V-portion of the pumping scheme). One advantage to this type of pumping design over simpler configurations, such as the V-pumping geometry, is that the additional passes allow more of the gain medium to be pumped.

The FIR cavity is about 2 m long and utilizes a nearly confocal mirror system, consisting of two 1.9 m radius-of-curvature concave gold-coated copper mirrors with a 50 mm diameter. Four Invar rods connect the end plates holding the FIR mirrors and the 2 m long, 59 mm inner diameter, Pyrex glass tube. One copper mirror is attached to a micrometer and is moved to tune the FIR cavity. The output coupling is varied by moving the 45° copper mirror radially in and out of the cavity mode. The generated FIR radiation is then sent through a polypropylene window, 0.634 μm thick, and focused by an off-axis parabolic mirror onto a metal-insulator-metal (MIM) point contact diode.

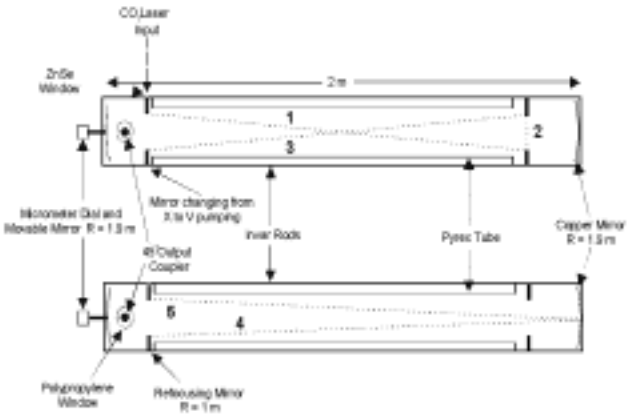

Figure 2
Side view of the optically pumped molecular laser system.

Wavelength measurements
Initial wavelength measurements of the FIR radiation were made by tuning the Fabry-Perot cavity with the movable end mirror and measuring the mirror displacement for 20 wavelengths of that laser mode. The value obtained is accurate to within ± 0.5 μm. A set of absorbing filters, calibrated by wavelength, discriminates against CO_2 laser radiation reaching the detector as well as helping distinguish different FIR wavelengths. The relative polarizations of the FIR emissions with respect to the CO_2 laser lines were measured with a multi-Brewster-angle polarization selector.

Frequency Measurements
FIR laser frequencies were measured using the three-laser heterodyne technique discussed elsewhere. [9,10] In general, different but known frequencies are mixed together to produce a sum or difference frequency. This frequency is then combined with a signal of unknown frequency. A "beat" , analogous to the beat heard between two musical tones, between the unknown and the sum or difference frequency can then be observed on a spectrum analyzer. If the separation between these frequencies is greater than the range of the spectrum analyzer, a microwave source may be added to decrease the gap.

The experimental setup is shown in Fig. 3. Two CO_2 laser frequencies were combined to create a difference frequency in the FIR region. The particular lines chosen to generate the difference frequency were based on the wavelength measurement of the unknown FIR emission. These CO_2 frequencies were stabilized by locking each laser to a saturation dip in the 4.3 μm fluorescence signal from an external reference cell.

The beat note, monitored by means of a spectrum analyzer, shown in Fig. 4, is used to determine the unknown frequency ν_x through the relation:

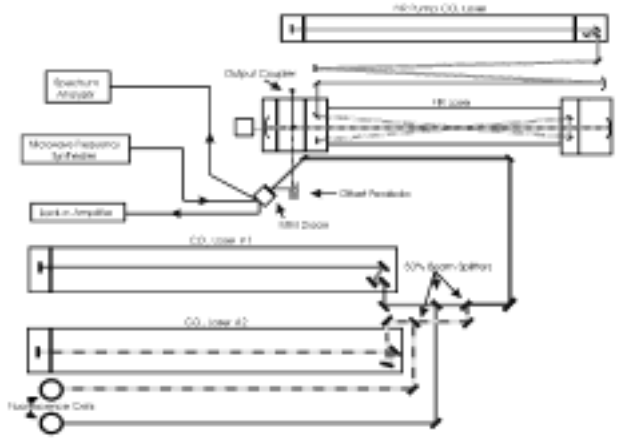

Figure 3
Diagram of the FIR laser and the frequency measurement setup.

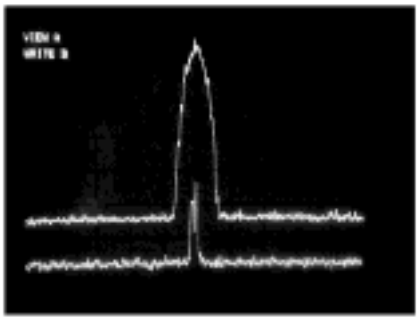

Figure 4
Spectrum analyzer display of the time-averaged beat note between the difference frequency (generated by the 9R(18) and 10R(30) CO_2 laser lines), the microwave source and the unknown FIR frequency. The center frequency is 2 850 703.9 MHz.

$$\nu_x = n_1 \nu_{CO2} \pm n_2 \nu_{\mu w} \pm \nu_{beat} , \quad (1)$$

where ν_{CO2} is the difference frequency synthesized by two CO_2 lasers, $\nu_{\mu w}$ the microwave frequency, and ν_{beat} the beat frequency. The integers, n_1 and n_2, define the mixing order of the difference and microwave frequencies, respectively (first-order, second-order, etc.).

A MIM point contact diode was used as a harmonic mixer, combining the signals from the laser and microwave sources. The signal from the MIM diode was fed into a preamplifier connected to a spectrum analyzer [11] to measure the intermediate frequency beat note by comparison with a marker generated by a synthesizer [11]. When necessary, a microwave source[11], operating between 0 and 18 GHz was used. The values of n_1 and n_2, as well as the ± sign in Equation 1 are determined experimentally by either tuning the FIR laser cavity or by increasing (or decreasing) the microwave frequency slightly to get a small shift in the beat note on the spectrum analyzer.

The one-sigma uncertainty of frequency measurements is $\Delta\nu/\nu = 2 \times 10^{-7}$. It is due mainly to the uncertainty in the setting of the FIR laser cavity to the center of its gain curve. For minimizing this uncertainty, we tuned the FIR laser across its gain curve and observed the change to the beat note on the spectrum analyzer using a peak hold feature. The value of this frequency is calculated from the average of ten measurements recorded with varying microwave frequencies. In addition, these measurements were made with at least two different sets of CO_2 laser lines.

DATA PRESENTATION AND INTERPRETATION OF RESULTS

In Table 1, we show the five new frequencies measured, arranged in order by their pump lines. The wavelengths and wavenumbers were calculated from the average frequency using 1 cm^{-1} = 29 979.2458 MHz. All FIR emissions were observed to have parallel (||) polarizations with respect to the CO_2 laser. With the laser emission from the 10R(32) pump line, two peaks were observed.

CO_2 Pump	Frequency (MHz)	Wavelength (μm)	Wavenumber (cm^{-1})
9R(6)	4 413 279.1	67.930	147.2111
10R(32)	3 582 585.2	83.680	119.5022
10R(32)	3 582 579.9	83.680	119.5020
10R(16)	3 463 216.2	86.565	115.5205
10P(32)	2 850 703.9	105.164	95.0892

Table 1
New frequencies measured for the laser emissions from optically pumped CD_3OH. All FIR emissions have their polarization parallel to those of the CO_2 pump lines.

These peaks, separated by approximately 5.3 MHz, may indicate a doublet or an experimental artifact based on the laser cavity used. This includes, but is not limited to, the possibility of observing a higher order TEM mode, or by pumping the FIR medium on the edge rather than in the center of its gain curve. The existence of a doublet only can be proven by determining its spectroscopic assignment. Experimental verification could be found by observing this line in another type of FIR cavity. The FIR frequencies were measured for the first time in this work under optimal operating conditions. A slight shift in frequency (possibly a few MHz) may still occur due to the type of FIR cavity and pumping geometry used. [8, 12, 13]

The expansion of frequency measurement techniques from the microwave to the optical regions has made a significant impact on the fields of metrology and spectroscopy. Despite the uncertainties, the FIR frequencies measured here represent an increase in accuracy over the calculated frequencies for CD_3OH. The frequencies measured using this OPML system, with the X-V pumping geometry, will be useful for future assignments of FIR laser emissions from this molecule by calculation of combination loops from high-resolution Fourier transform data. [14] These results can also be used for laser Stark and laser magnetic resonance spectroscopy where OPML emissions serve as a source of strong coherent FIR radiation. Finally, the information gained from these frequencies will help provide a more complete picture of CD_3OH in the far-infrared region.

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