Frequency Measurement of FIR Laser Emissions From Optically Pumped CH_3OD

Paul Noffke

Faculty Sponsor: Michael Jackson, Department of Physics

ABSTRACT

A three-laser heterodyne frequency measurement system has been developed to measure far-infrared (FIR) laser frequencies with fractional uncertainties up to $\pm 2 \times 10^{-7}$. Seven new frequencies have been measured for previously observed FIR laser emissions of optically pumped CH_3OD. The frequencies and wavelengths of these lines are reported for emissions ranging from 80.1 to 183.3 μm. A previously measured FIR laser emission from CH_3OD, observed at 57.2 μm, was also re-measured in this work.

INTRODUCTION

A significantly improved optically pumped molecular laser (OPML) system was recently developed to generate short-wavelength ($\lambda < 150$ μm) FIR laser emissions [1]. Since then, over fifty-five laser emissions have been discovered from several optically pumped methanol isotopes (CD_3OH, $^{13}CH_3OH$, $^{13}CD_3OD$, CHD_2OH, CH_2DOH, CH_3OD and $^{13}CD_3OH$) and hydrazine (N_2H_4) with the wavelengths ranging from 26.3 to 183.3 μm [1-6]. The objective of this work was to develop a frequency measurement system to effectively measure these recently discovered as well as other previously observed short-wavelength laser emissions.

The partially deuterated methanol isotope, CH_3OD, which has been the subject of several recent studies [5, 7-9], was selected for reinvestigation because it has been found to produce over 180 laser emissions in the FIR, with about forty-five percent of these emissions having wavelengths shorter than 150 μm [5, 7-11]. In this paper, we discuss the experimental setup of the OPML and three-laser heterodyne frequency measurement systems as well as report the measured frequencies of eight optically pumped FIR laser emissions from CH_3OD.

EXPERIMENTAL DETAILS

The experimental apparatus used to search for FIR laser lines (described in detail elsewhere [1, 3]) consisted of a tunable Fabry-Perot cavity optically pumped with a X-V pumping geometry by a continuous-wave carbon dioxide (CO_2) laser. The FIR cavity utilized a nearly confocal mirror system with one end mirror mounted on a micrometer to tune the cavity into resonance with the FIR laser radiation. Laser wavelengths were measured by scanning over 20 adjacent longitudinal modes for a particular laser emission.

The FIR laser frequencies were measured using the three-laser heterodyne technique discussed in detail in Refs. [12, 13]. 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 between the unknown and the known 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 for this system is shown in Fig. 1. Here, 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 laser emission. These CO_2 reference frequencies were stabilized by locking each laser to a saturation dip in the 4.3 μm fluorescence signal from an external reference cell.

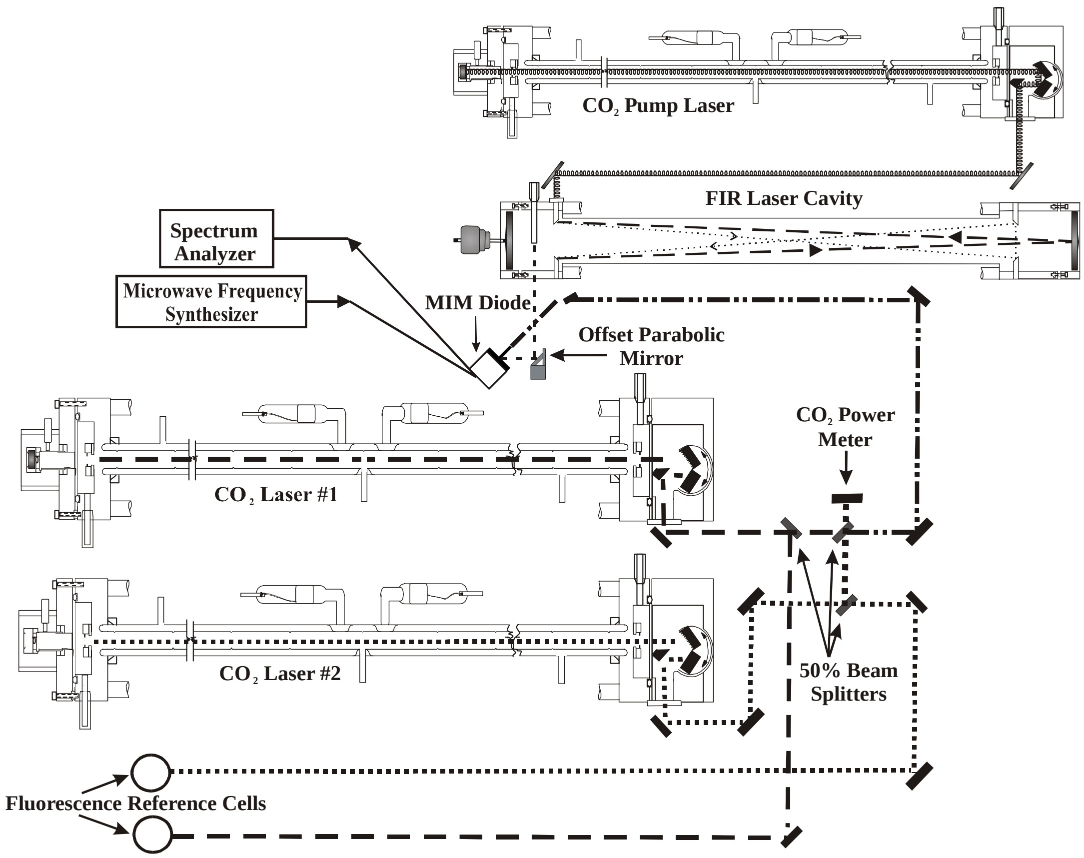

Figure 1. The OPML and three-laser heterodyne systems.

The beat note, monitored by means of a spectrum analyzer and shown in Fig. 2, is used to determine the unknown laser frequency, ν_{FIR}, through the relation:

$$\nu_{FIR} = |n_1\nu_{CO2(I)} - n_2\nu_{CO2(II)}| \pm m\nu_{\mu w} \pm \nu_{beat} \qquad (1)$$

where $|n_1\nu_{CO2(I)} - n_2\nu_{CO2(II)}|$ 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, n_2, and m define the harmonics of the difference and microwave frequencies, respectively (first-order, second-order, etc.). A metal-insulator-metal (MIM) point contact diode was used as a harmonic mixer, combining the signals from the lasers and microwave source. The signal from the MIM diode was fed into an Avantek AWL-1200B amplifier connected to a HP8558B spectrum analyzer, with a 0.1 – 1.5 GHz range, to measure the intermediate frequency beat note by comparison with a marker generated by a standardized HP8640B frequency signal generator. When necessary, a HP8675A microwave source, operating between 2 and 18 GHz, was used. The values of n_1, n_2, m and the ± sign in Eq. (1) were determined experimentally by either tuning the FIR laser cavity or by increasing (or decreasing) the microwave frequency slightly in order to get a small shift in the beat note on the spectrum analyzer.

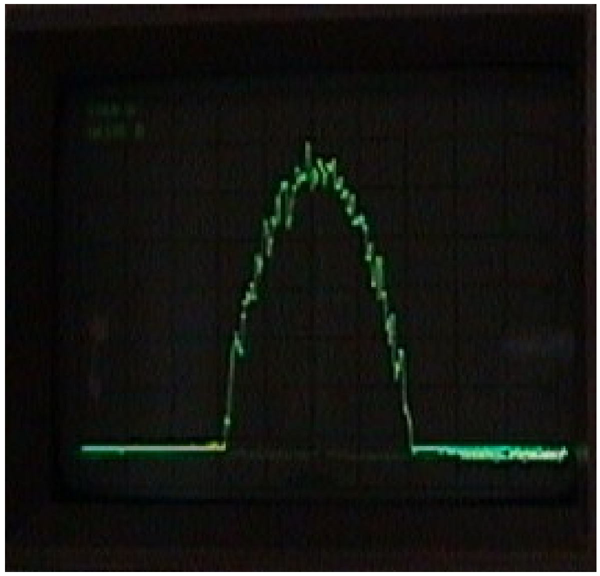

Figure 2. Spectrum analyzer display of the time-averaged beat note between the difference frequency (generated by the 9P12 and the 10R26 CO_2 laser lines), the microwave source and the unknown FIR frequency for the 135.2 m wavelength from optically pumped CH_3OD.

The one-sigma uncertainty of frequency measurements is $\Delta\nu/\nu = \pm 2 \times 10^{-7}$. 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 was calculated from the average of at least ten measurements recorded with varying microwave frequencies. In addition, these measurements were made with at least two different sets of CO_2 reference laser lines.

RESULTS

Table 1 lists seven new measurements of FIR laser frequencies and one re-measured FIR laser frequency, arranged in order by their CO_2 pump lines. The wavelengths and wavenumbers were calculated from the average frequency using 1 cm^{-1} = 29 979.2458 MHz. Although this system successfully reproduced seven known FIR laser frequencies belonging to CH_3OD, one emission, the 57.151 μm laser line generated using the 9R8 CO_2 pump, was not in agreement. The frequency of this laser line was originally reported as 5 245 612.9 ± 1.5 MHz [13] but was measured in the present work to be 5 245 609.8 ± 1.1 MHz. It is our opinion that the latter frequency is the better of the two, having been measured twenty-six times using three different sets of CO_2 reference laser lines. The slight difference between the two measured values might be due to an asymmetric gain curve observed in Ref. [15]. When such curves were observed in the present work, an iris in each of the laser cavities (the CO_2 pump laser, FIR laser and CO_2 reference lasers) was used to eliminate higher order modes and reshape the gain curve to a symmetric pattern, typical of a fundamental laser mode. Finally, several recently discovered emissions (the 54.0 and 90.1 μm lines [5]) were observed in this work, but due to insufficient FIR laser power, their frequencies were not measured.

CONCLUSIONS

Despite the uncertainties, the FIR laser frequencies measured in this work for optically pumped CH_3OD represent an increase in accuracy over those calculated using their measured wavelengths. These frequencies will be useful for confirming spectroscopic assignments of FIR laser emissions by calculation of combination loops using high-resolution Fourier transform data [21]. The accurate measurement of FIR laser emissions will also permit these lines to be used as sources of strong, coherent FIR radiation for laser Stark and laser magnetic resonance spectroscopy. Finally, the information gained from these frequencies will help provide a more complete picture of CH_3OD in the far-infrared region.

Table 1. New frequency measurements from optically pumped CH_3OD

Pump	λ (μm)	ν (MHz)	ν (cm^{-1})	Reference
9R8	57.151	5 245 609.8 ± 1.1	174.9747	14, 15[a]
9P6	183.266	1 635 829.7 ± 0.6	54.5654	5
9P6	135.186	2 217 625.3 ± 0.7	73.9720	16
9P26	101.207	2 962 180.3 ± 0.9	98.8077	17
9P30	89.308	3 356 826.0 ± 0.9	111.9717	18
9P32	80.064	3 744 394.6 ± 1.0	124.8996	17, 19
9P32	88.553	3 385 448.7 ± 0.9	112.9264	20
10R42	140.909	2 127 560.5 ± 0.8	70.9678	17

[a] FIR laser frequency was previously measured

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