Measurement of Far-Infrared Laser Frequencies From Optically Pumped CHD_2OH

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ABSTRACT

A three-laser heterodyne system was used to measure the frequencies of eleven previously observed optically pumped far-infrared (FIR) laser emissions generated by the CHD_2OH methanol isotope. These newly measured frequencies have fractional uncertainties of $\pm 2 \times 10^{-7}$ and correspond to laser wavelengths ranging from 47.8 to 238.0 μm. The pump laser offset frequency was also measured for the first time for fifteen CHD_2OH FIR laser emissions.

INTRODUCTION

The partially deuterated methanol isotope, CHD_2OH, was first investigated as a FIR laser medium by Ziegler and Dürr in 1978 [1]. Following their discovery of 11 FIR laser emissions, Facin and co-workers [2] discovered an additional 82 FIR laser lines in 1989. In 2002, Moraes *et al.* [3], Evenson *et al.* [4] and Viscovini *et al.* [5] reported the discovery of 16, 6 and 5 FIR laser lines, respectively, which included the shortest (32.8 μm [4]) and longest (710.0 μm [5]) FIR laser emissions for this methanol isotope. All FIR laser emissions reported in Refs. [1-5] were found using a continuous-wave (cw) carbon dioxide (CO_2) pump laser. In 2003, a waveguide pulsed CO_2 pump laser was used to discover 12 FIR laser emissions [6]. Recently, the first measurement of FIR laser frequencies from optically pumped CHD_2OH was performed for eleven lines ranging in wavelength from 102.9 to 212.4 μm [7]. As with other methanol isotopes, CHD_2OH can serve as a source of strong, coherent FIR laser radiation. This isotope has 132 known laser emissions, 58 of which are in the short-wavelength portion of the FIR region (25 μm$\leq \lambda <$ 150 μm). In this paper, the experimental setup of the three-laser heterodyne system will be discussed and 11 FIR laser frequencies reported. In addition, the offset of the CO_2 pump laser from its center frequency will be reported for 15 FIR laser emissions.

EXPERIMENTAL DETAILS

The FIR laser consisted of a tunable Fabry-Perot cavity, optically pumped in an X-V geometry by a 2 m long cw CO_2 laser [8]. 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 traversing at least twenty adjacent longitudinal modes for a particular laser emission. The sample of CHD_2OH, 98% D_2 enriched, was obtained from Cambridge Isotope Laboratories.

The FIR laser frequencies were measured using the three-laser heterodyne technique, discussed in detail in Refs. [9-11]. Two CO_2 laser frequencies were combined in a Tungsten-Nickel metal-insulator-metal (MIM) point contact 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.

The spectrum analyzer used in this work was upgraded from an HP 8558B to an Agilent E4407B. The most significant improvement resulting from this upgrade was an increase in spectral coverage offered by the spectrum analyzer, from 1.5 GHz to 26.5 GHz. The Agilent E4407B spectrum analyzer also has an internal frequency standard, eliminating the need for an external signal generator to accurately measure the beat note.

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

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

where $| n_1 \nu_{CO2(I)} - n_2 \nu_{CO2(II)} |$ 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 (first-order, second-order, 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 note on the spectrum analyzer. Figure 1 shows a typical spectrum analyzer display with two observed beats between the 167.495 μm laser line of CHD_2OH (obtained using the 10*R*38 CO_2 pump), the difference frequency generated by the 9*P*16 and 10*R*46 CO_2 laser emissions and a 2430 MHz microwave frequency. The left beat (observed at ≈ 1118 MHz) required the use of a first-order microwave frequency (m = –1) while the right beat (observed at ≈ 1312 MHz) required the use of a second-order microwave frequency (m = –2).

The CO_2 reference lasers were frequency stabilized to their saturation dips in the 4.3 μm fluorescence signal from an external reference cell [12]. 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 reshape the gain curve to a symmetric pattern. The FIR laser frequency was calculated from the average of at least fifteen measurements recorded with varying microwave frequencies. In addition, these measurements were made with at least two different sets of CO_2 reference laser lines.

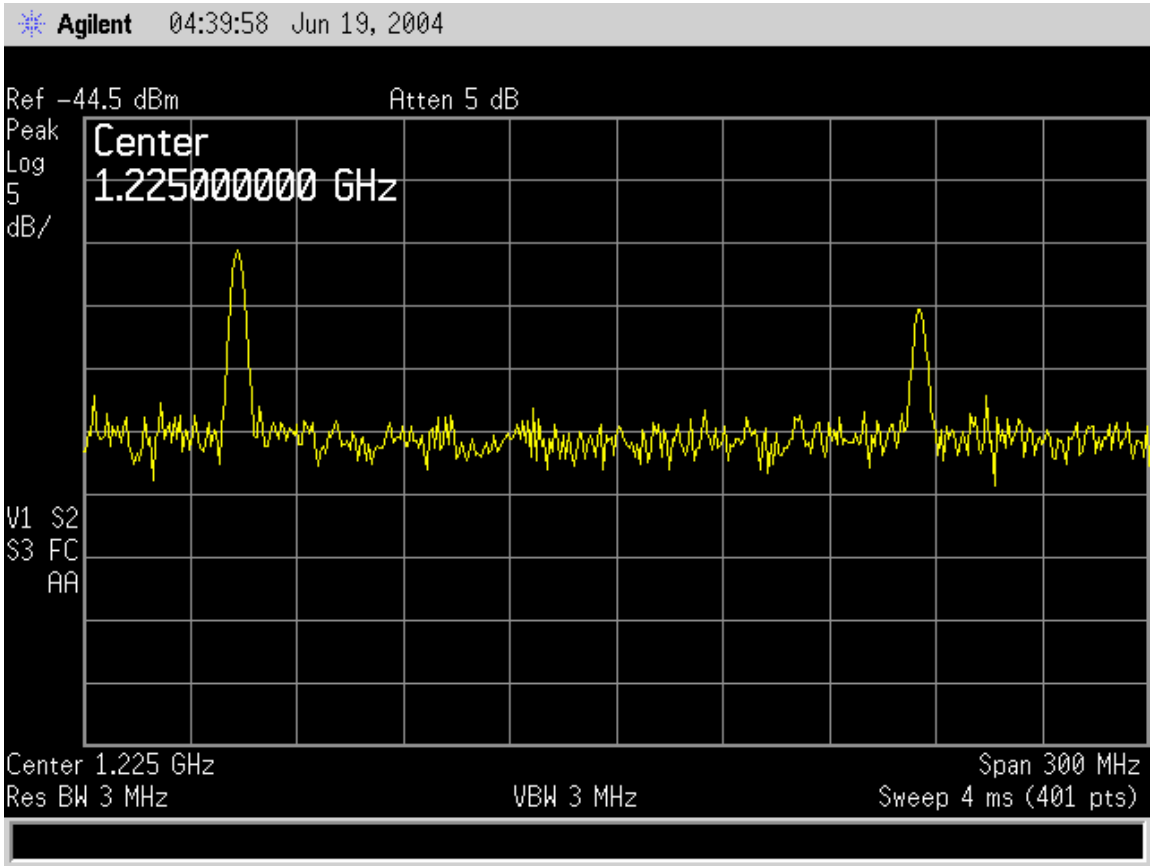

Figure 1. Two observed beats between the 167.495 μm laser line of CHD_2OH (obtained using the 10*R*38 CO_2 pump), the difference frequency generated by the 9*P*16 and 10*R*46 CO_2 laser emissions and a 2430 MHz microwave frequency.

Lastly, the pump offset from the CO_2 line center (the difference between the CHD_2OH 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 same CO_2 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).

RESULTS

The measured FIR laser frequencies from optically pumped CHD_2OH 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. Although the FIR laser frequencies were measured under optimal operating conditions, a slight shift in frequency (possibly a few MHz) may still occur due to the type of CO_2 pump laser, FIR cavity and pumping geometry used [13].

Table I. Measured Laser Frequencies generated by the Optically Pumped CHD_2OH Methanol Isotope

CO_2 Pump	Wavelength (μm)	Frequency (MHz)	Wavenumber (cm^{-1})	Offset [b] (MHz)	Ref.
9*R*38	115.283	2 600 484.8 ± 0.6[a]	86.7428	0	2
9*R*30	134.424	2 230 205.7 ± 0.5[a]	74.3917	+19	2
9*R*18	140.412	2 135 087.2 ± 0.5	71.2188	–9	3
	164.332	1 824 313.4 ± 0.4	60.8525	+9	1
9*R*16	217.710	1 377 026.2 ± 0.3	45.9327	+16	2
10*R*44	157.280	1 906 104.3 ± 0.4	63.5808	–9	4
10*R*40	79.987	3 748 012.6 ± 0.8	125.0202	–18	2
10*R*38	53.108	5 644 974.7 ± 1.2	188.2961	–9	4
	167.495	1 789 860.7 ± 0.4	59.7033	–3	1
10*R*16	47.825	6 268 533.1 ± 1.3	209.0958	+22	4
	55.512	5 400 525.5 ± 1.1	180.1421	–27	3
	178.285	1 681 532.9 ± 0.4	56.0899	–27	1
10*P*16	102.906	2 913 263.4 ± 0.6[a]	97.1760	–13	2
10*P*18	212.406	1 411 410.5 ± 0.4[a]	47.0796	+5	2
	238.040	1 259 421.6 ± 0.3	42.0098	+9	1

[a] Previously reported FIR laser frequency [7].
[b] Measured to within ±10 MHz.

CONCLUSIONS

In this work, eleven FIR laser frequencies and fifteen offset frequencies for the CHD_2OH methanol isotope were measured for the first time. The frequencies measured in this work represent an increased accuracy over those calculated using the measured wavelengths and thus will be useful for future assignments of FIR laser emissions by calculation of combination loops from high-resolution Fourier transform data [14]. Along with helping to provide a more complete picture of CHD_2OH in the far-infrared region, these emissions can serve as sources of strong, coherent FIR radiation for a variety of spectroscopic techniques, including laser Stark and laser magnetic resonance spectroscopy.

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