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Discovery and measurement of optically pumped far-infrared laser emissions in $^{13}CD_3OH$

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ABSTRACT We report the discovery of four laser emissions from the partially deuterated C-13 methanol isotope $^{13}CD_3OH$ when optically pumped with a cw carbon dioxide (CO_2) laser. The wavelengths of these lines, ranging from 45.3 to 108.9 μm, are reported along with their polarizations relative to the CO_2 pump laser, operating pressure, and relative intensity. A three-laser heterodyne system was then used to measure the frequencies of eight optically pumped laser emissions from this methanol isotope. These emissions range from 53.4 to 126.1 μm and are reported with fractional uncertainties up to $\pm 2 \times 10^{-7}$.

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1 Introduction

Since the first successful generation of far-infrared (FIR) laser radiation from $^{13}CD_3OH$ in 1984 [1], this species has produced 189 laser lines, ranging from 32 to 2615 μm when optically pumped by CO_2 lasers [2], with approximately one hundred in the short-wavelength ($\lambda < 150$ μm) portion of the FIR. This includes the FIR laser emission at 127 μm, a very efficient line [2, 3], which together with the 123.5- and 118.8-μm laser lines from CH_3OH form a triad of the most efficient optically pumped laser emissions known. In addition, of the 189 laser emissions generated from $^{13}CD_3OH$, the frequencies of eighty lines have been accurately measured by heterodyne techniques and one hundred have been spectroscopically assigned [2].

In this communication, we report the measurement of eight FIR laser frequencies belonging to $^{13}CD_3OH$ as optically pumped by six different CO_2 laser lines. Frequencies were measured with the three-laser heterodyne technique by use of a recently designed short-wavelength optically pumped molecular laser (OPML) system to generate the FIR laser emissions. During the process of these measurements, four FIR laser emissions were also discovered. The wavelengths of two of these new lines were recorded without a corresponding frequency measurement.

2 Experimental details

A recently designed OPML system, consisting of a CO_2 pump laser [4] and a FIR laser cavity [5], was used to generate FIR laser emissions. The $^{13}CD_3OH$ laser medium was excited by redirecting the CO_2 radiation by means of an X–V pump geometry, recently shown to successfully stimulate short-wavelength FIR laser emissions [5–8]. Laser wavelengths were measured by scanning over 20 adjacent longitudinal modes for a particular laser emission. Using this method, we measured FIR laser wavelengths with an uncertainty of ±0.5 μm. The relative polarizations of the FIR laser emissions with respect to the CO_2 laser lines were measured with a multi-Brewster-angle polarization selector. The intensities of the laser lines were measured with a pyroelectric detector using various filters. The filters served to attenuate any scattered CO_2 laser radiation directed through the FIR laser output window as well as help distinguish different FIR wavelengths [5]. The $^{13}CD_3OH$ (99% ^{13}C-enriched and 98% D-enriched) sample was obtained from Cambridge Isotope Laboratories.

The frequencies of OPML emissions were measured using the three-laser heterodyne technique discussed in detail in [8] and [9]. Here, two CO_2 laser frequencies were combined to create a difference frequency in the FIR region. This difference frequency was beat with the unknown FIR laser frequency, ν_{FIR}, and the particular lines chosen to generate the difference frequency were based on the wavelength measurement of the unknown FIR laser 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, generated in a metal–insulator–metal (MIM) point-contact diode, was monitored by means of a spectrum analyzer and used to determine ν_{FIR} through the relation

$$\nu_{FIR} = |n_1 \nu_{CO_2(I)} - n_2 \nu_{CO_2(II)}| \pm m \nu_{\mu wave} \pm \nu_{beat}, \quad (1)$$

where n_1, n_2, and m are experimentally determined integers that correspond to the respective harmonics (first-

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order, second-order, etc.) generated in the MIM diode; $\nu_{\mu\text{wave}}$ is the microwave frequency (when necessary), and ν_{beat} is the beat frequency. The fractional uncertainty of a frequency measurement is at least $\Delta\nu/\nu = \pm 2\times 10^{-7}$ and is due mainly to the uncertainty in the setting of the FIR laser cavity to the center of its gain curve.

3 Results and discussion

The newly discovered FIR laser emissions from optically pumped $^{13}CD_3OH$ are listed in Table 1 and are arranged in order of their CO_2 pump line. Along with their wavelengths, their polarizations with respect to the CO_2 pump laser and their operating pressures and relative intensities are also reported. The relative intensity of the FIR output is labeled as either M or W corresponding to ranges in power between 0.1–0.01 mW and 0.01–0.001 mW, respectively.

Table 2 gives the measured frequencies for eight laser emissions from optically pumped $^{13}CD_3OH$. The value of each frequency is an average of 10 measurements recorded with varying microwave frequencies (when necessary) by use of two different sets of CO_2 laser lines. The frequency for the 108.891-µm line was an average of 20 measurements. The wavelengths and wavenumbers, listed in Table 2, were calculated from the average frequency using $1\ \text{cm}^{-1} = 29\,979.2458$ MHz. The FIR laser frequencies were measured under optimal operating conditions. A slight shift in frequency may still occur due to the type of FIR cavity and pumping geometry used [13].

CO_2 Pump	Wavelength (µm)	Rel. Pol.	Pressure (Pa)	Rel. Int.
10*R*34	68.8	∥	20.0	W
10*R*22	45.3	∥	16.0	M
	108.891[a]	∥	26.6	M
10*P*12	103.318	∥	33.3	W

[a] The 110 µm line previously reported [1] was not observed in this work

TABLE 1 New FIR laser emissions from optically pumped $^{13}CD_3OH$

CO_2 Pump	Wavelength (µm)	Frequency (MHz)	Wavenumber (cm^{-1})
9*P*10	72.858	4 114 772.2 ± 0.9	137.2540[a]
	126.164	2 376 217.0 ± 0.5	79.2621[a]
10*R*24	81.717	3 668 687.3 ± 1.5	122.3742[b]
10*R*22	108.891	2 753 148.1 ± 0.6	91.8351[c]
10*R*8	97.625	3 070 869.7 ± 0.7	102.4332[d]
10*P*10	53.389	5 615 302.4 ± 1.2	187.3063[e]
10*P*12	84.630	3 542 371.0 ± 0.8	118.1608[e]
	103.318	2 901 641.9 ± 0.6	96.7884[c]

[a] Wavelength first reported in ref. [1]
[b] Wavelength first reported in ref. [10]
[c] New FIR laser emission
[d] Wavelength first reported in ref. [11]
[e] Wavelength first reported in ref. [12]

TABLE 2 New measurements of FIR laser frequencies from $^{13}CD_3OH$

4 Conclusions

We reported the discovery of four laser emissions from optically pumped $^{13}CD_3OH$ and the measurement of eight FIR laser frequencies from this methanol isotope. The new laser emissions will be useful for filling the gaps currently existing in the short-wavelength portion of the FIR region. Due to the accuracy with which the laser frequencies were measured, this work will be useful for future assignments of FIR laser emissions by calculation of combination loops from high-resolution Fourier-transform data.

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