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NEW FAR INFRARED LASER LINES OBTAINED BY OPTICALLY PUMPING $^{13}CD_3OD$

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We have obtained laser action on 34 far infrared lines for the first time in fully deuterated methyl alcohol with the ^{13}C isotope ($^{13}CD_3OD$), and we have measured the frequency of 13 lines. The molecule was pumped by a cw CO_2 laser. We have measured the wavelength, the relative polarization, the relative intensity of most lines, the frequency, and the CO_2 pump frequency offset of the strongest lines. The new lines are distributed in the wavelength region from 75.27 µm to 464.7 µm.

Key Words: $^{13}CD_3OD$, laser frequency measurement, FIR laser, new laser lines, CO_2 laser, wavelengths.

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INTRODUCTION

It is well known that the methyl alcohol molecule (CH_3OH) is the active medium which has generated the great majority of known FIR laser lines when optically pumped by CO_2 lasers. Although the substitution of deuterium for hydrogen and of ^{13}C for ^{12}C in the molecule shifts its infrared spectrum, it still falls within the 900-1100 cm-1 pumping range of the CO_2 laser. Hence many new FIR laser lines can be obtained by pumping the various isotopic species with these lasers. For example, the completely deuterated methyl alcohol molecule (CD_3OD) produces up to 140 new laser lines in the wavelength range from 35 µm to 2.9 mm [1,2].

For the present work we explore for the first time the $^{13}CD_3OD$ molecule in search of new submillimeter wave laser lines.

EXPERIMENT

We have pumped the molecule with lasing lines from a 2m long, 10mm diameter, cw, waveguide CO_2 laser which oscillates from $R_{II}(38)$ to $R_{II}(6)$, $P_{II}(6)$ to $P_{II}(44)$, $R_I(40)$, to $R_I(6)$ and $P_I(6)$ to $P_I(38)$ delivering powers of 45W on the strongest lines, and which tunes about 70 MHz on each line. Also, we have used some of the pumping lines from a 1m long, 5mm bore diameter cw-waveguide CO_2 laser with powers up to 25W on the strongest lines. The later CO_2 laser oscillates in all the lines of the following ranges: $R_{II}(50)$ to $R_{II}(2)$, $P_{II}(2)$ to $P_{II}(52)$, $R_I(48)$ to $R_I(4)$ and $P_I(2)$ to $P_I(56)$ and tunes about 120 MHz on each line. In each laser, the partially transmitting IR mirror was PZT driven for fine tuning of the laser over its gain curve.

The FIR cavity described elsewhere[(3)] consisted of an open structure (non-waveguide) resonator 104 cm long with 2 m radius of curvature concave mirrors at each end. One mirror was mounted on a micrometer to tune the resonant frequency of the cavity and the other one was fixed. The laser tube consisted of a 1m long, copper cylinder 51 mm in diameter. Transverse pumping was achieved by bringing the CO_2 radiation through a side window into the tube at an angle of 75° with respect to the laser axis. Longitudinal pumping was done through a 1mm hole centered at the fixed mirror. The FIR radiation was reflected out of the cavity by a variable coupler which consisted of a 45° mirror 6 mm in diameter whose distance from the laser axis was adjustable. The FIR radiation then exited through a polyethylene window located at the side of the cavity and was detected with a metal-insulator-metal (MIM) point contact diode. The FIR cavity was provided with a microphone. The generated photoacoustic signal was used to tune the CO_2 laser frequency to the methyl alcohol absorption.

The wavelengths were determined to $\pm$ 0.1 μm by measuring a number of half wavelengths in a measured scan of the laser end mirror of approximately 5 mm.

The frequencies were measured by synthesizing each FIR frequency from the difference between two saturated-absorption, fluorescence-stabilized CO_2 lasers as already reported.(4) The MIM point contact diode served as the mixer.

The pumping CO_2 frequency offset for each line was measured by heterodyning the pumping radiation with one of the actively stabilized CO_2 lasers retuned to the pump line.

The polarizations of the FIR lines relative to the CO_2 laser lines were determined by using a Brewster angle stacked plate polarizer in front of the FIR detector.

Pressures were measured with a thermocouple gauge calibrated with a capacitance manometer and the values presented in Table I were adjusted to obtain maximum FIR output power.

EXPERIMENTAL RESULTS AND COMMENTS

We have measured for the first time laser action in the $^{13}CD_3OD$ molecule on 34 lines in the wavelength range from 75.27 μm to 464.7 μm.

Table I lists the $^{13}CD_3OD$ laser lines along with their relative polarization, the pressure for maximum FIR power, the relative FIR intensity and the corresponding CO_2 pump lines.

Table II lists the wavelengths, the measured frequencies, the vacuum wavenumbers, the pressures, the CO_2 pump lines and the CO_2 frequency offsets of the frequency measured FIR $^{13}CD_3OD$ laser lines.

The λ = 148.62 μm laser line pumped by $R_I(30)$ has a wavelength value close to the λ = 150 μm laser line reported by Herman et al. on $^{12}CD_3OD$[5] and both have the same polarization. Since we have not observed such a line in our previous work on $^{12}CD_3OD$[6] using essentially the same FIR cavity, we believe that the λ = 148.62 µm laser line is a different line from the one reported by Herman et al. and it is generated from the $^{13}CD_3OD$ isotopic species.

Fifteen out of a total of 36 FIR laser lines reported on $^{13}CD_3OH$[7] have also lased with $^{13}CD_3OD$, however they were attributed to $^{13}CD_3OH$ because they were observed to be weaker in the completely deuterated methyl alcohol. Either because of hydrogen - deuterium exchange on the wall or the fact that our $^{13}CD_3OD$ was 99% $^{13}CD_3OD$ and 1% $^{13}CD_3OH$, it was difficult to separate $^{13}CD_3OH$ lines from $^{13}CD_3OD$ lines.

It was also noteworthy that FIR laser lines in $^{13}CD_3OD$ were more critically dependent on pressure and CO_2 PZT tuning than in other isotopes of methyl alcohol.

Finally, we believe that more laser lines can still be found in $^{13}CD_3OD$ with improvements in the CO_2 coupling to the FIR cavity, a wider variation in the pressure, and with the use of a larger bore FIR cavity suitable to accomodate larger wavelength modes.

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Table I - Summary of New Laser Lines in $^{13}CD_3OD$

CO_2 Pump Line	$^{13}CD_3OD$ Laser Line[a] (μm)	Relat. Polar.	Press. for Maximum Pa(mTorr)[b]	Relative FIR Intens.	CO_2 Power (W)
$R_{II}(34)$	82.1	//	18(135)	200	31.5
$R_{II}(32)$	150.3	//	18(135)	100	28.5
$R_{II}(28)$	227.0	⊥	17(125)	400	35
$R_{II}(26)$	353.1*	⊥	17(125)	1200	22
$R_{II}(24)$	75.27*	//	24(180)	600	27.5
$R_{II}(22)$	175.1	//	12(90)	50	33
$R_{II}(20)$	241.6	//	14(105)	250	35.5
"	417.3	//	14(105)	200	33
$R_{II}(14)$	118.55*	//	17(125)	400	35
"	353.3	//	10(75)	800	36.5
"	82.4	⊥	18(135)	400	35
$R_{II}(10)$	75.5	⊥	12(90)	60	28
$P_{II}(12)$	126.2	//	12(90)	500	27
"	247.0	//	12(90)	200	31
$P_{II}(24)$	151.0	//	12(90)	100	30.5
$P_{II}(28)$	151.8	//	12(90)	70	32
"	407.1	//	7(55)	250	32.5
$P_{II}(32)$	243.0	//	8(60)	60	31.5
"	358.4	//	8(60)	250	31.5
$P_{II}(38)$	129.2	⊥	9(65)	120	20
$R_{I}(30)$	148.62*	//	9(65)	250	24.5

Table I - Continued

R_I (26)	272.96*	⊥	14(105)	200	22
R_I (20)	173.64	⊥	18(135)	4000	41
R_I (16)	109.93	//	9(65)	400	41
"	109.94	//	9(65)	400	41
"	128.10	//	13(100)	150	41
"	93.6		12(90)		41
R_I (14)	324.14	⊥	8(60)	200	42.5
R_I (12)	209.23	⊥	6(45)	40	40
"	321.41	⊥	3(25)	50	40
R_I (8)	464.7	//	9(65)	600	27
P_I (12)	84.4*		12(90)	10	25
P_I (24)	124.25	//	20(155)	1000	25
"	216.36*	⊥	24(180)	120	26.5

[a]Two decimal figures are reported for lines whose frequencies have been directly measured (See Table II).

[b]Pressure at which each frequency was measured as determined by a thermocouple gauge calibrated with a capacitance manometer.
1 Torr = 133.3 Pa

*Lines which have been measured with the 104 cm long cw-waveguide laser.

Table II - Summary of the $^{13}CD_3OD$ Frequency Measurements

$^{13}CD_3OD$ Laser Line λ(μm)	Meas.Freq. (MHz) (Uncertainty: = ± 2x10^{-7})[a]	Vacuum Wavenumber (cm^{-1})[b]	$^{13}CD_3OD$ Pressure Pa(mTorr)	CO_2 Pump Line	CO_2 Freq. Offset (MHz)[c]
75.27	3 982 631.1	132.846 274	24(180)	$R_{II}(24)$	+13.8
109.93	2 727 211.7	90.969 992	10(75)	R_I (16)	+ 4.6
109.94	2 726 923.5	90.960 376	10 (75)	R_I (16)	+ 4.6
118.55	2 528 772.8	84.350 780	10 (75)	$R_{II}(14)$	- 3.3
124.25	2 412 757.9	80.480 942	24 (180)	P_I (24)	+ 5.7
128.10	2 340 291.8	78.063 731	13 (100)	R_I (16)	+ 4.9
148.62	2 017 218.5	67.287 167	9 (65)	R_I (30)	- 3.3
173.64	1 726 548.5	57.591 459	17(125)	R_I (20)	+ 2.0
209.23	1 432 817.8	47.793 659	7(55)	R_I (12)	-10.5
216.36	1 385 646.1	46.220 179	30(225)	P_I (24)	- 0.4
272.96	1 098 307.9	36.635 609	12(90)	R_I (26)	-19.7
321.41	932 741.3	31.112 901	3(25)	R_I (12)	+ 1.3
324.14	924 885.8	30.850 871	8(60)	R_I (14)	-14.9

[a]Estimated $\Delta\nu/\nu$ uncertainty in the reproducibility of the FIR laser frequency.

[b]Calculated from the measured frequency with c = 299 792 458 m/s.

[c]Uncertainty: ± 3MHz in the CO_2 Frequency Offset Measurement.

REFERENCES

1. S. Kon, T. Kachi, Y. Tsunawaki and M. Yamanaka, "Optically Pumped Far-Infrared Laser Lines in Deuterated Methyl Alcohol, CH_3OD, CH_2DOH, CHD_2OH, CD_3OH, CD_3OD, Reviews of Infrared and Mill. Waves, Vol. 2, 159-192 (1984), Ed. by Kenneth J. Button, M. Inguscio and F. Strumia.

2. A. Scalabrin, D. Pereira, E.C.C. Vasconcellos, K.M. Evenson, F.R. Petersen and D.A. Jennings, "Measurements of New FIR Lines in CD_3OD" 1984 IR & MM Waves Conference, Takarazuka Japan, pp. 505-506.

3. E.C.C. Vasconcellos, J.C. Wyss, F.R. Petersen and K.M. Evenson, "Frequency Measurements of Far Infrared CW Lasing Lines in Optically Pumped $CHCl_2F$", Int. J. Infr. & Mill. Waves, 4, 401-406 (1983).

4. F.R. Petersen, K.M. Evenson, D.A. Jennings, J.S. Wells, K. Goto, and J.J. Jimenez, "Far Infrared Frequency Synthesis with Stabilized CO_2 Lasers: Accurate Measurements of the Water Vapor and Methyl Alcohol Laser Frequencies", IEEE J.Q. Electr. QE-11, 838-843 (1975).

5. Herman H. and Prewer B.E., "New FIR Laser Lines from Optically Pumped Methanol Analogues", Appl. Phys. 19, 241-242 (1978).

6. E.C.C. Vasconcellos, A. Scalabrin, F.R. Petersen and K.M. Evenson, "New FIR Laser Lines and Frequency Measurements in CD_3OD", Int. J. Infr. & Mill. Waves, 2, 533-539 (1981).

7. M. Inguscio, K.M. Evenson, F.R. Petersen, F. Strumia, and E.C.C. Vasconcellos, "A New Efficient Far Infrared Lasing Molecule: $^{13}CD_3OH$, J. Infr. & Mill. Waves, 5, 1289-1296 (1984).