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A NEW EFFICIENT FAR INFRARED LASING MOLECULE: $^{13}CD_3OH$[1]

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Thirthy-six new cw laser lines ranging from 52 to 469 μm are obtained by pumping for the first time the isotope of methyl alcohol $^{13}CD_3OH$. The new laser line at 127.0 μm is one with the highest efficiency ever reported. Direct frequency measurements are reported for eleven new lines.

Introduction

Methyl alcohol was one of the first molecules which was optically pumped by CO_2 lasers to produce CW FIR laser radiation (1). At the present time 384 FIR laser lines have been observed to lase in normal CH_3OH pumped by the bands of

(+) Dr. F.R. Petersen passed away during the final preparation of this manuscript. We authors would like to dedicate this paper to our close friend and colleague.

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the normal or isotopically substituted or sequence CO_2 laser (2). More than 400 FIR lines are added to the list if $^{13}CH_3OH$ or CH_3OD, CD_3OD, CH_2DOH (4,5) isotopic methyl alcohol molecules are considered. The success of methyl alcohol as an active FIR medium derives from the excellent overlap which exists between the strongly absorbing C-O stretch band and the CO_2 laser emissions and from the complexity of the rotational spectrum. Furthermore the contribution of the internal rotation (OH group torsion) to the transition energy makes possible the generation of very short wavelength lines. As a matter of fact, laser lines at wavelengths shorter than 90 μm have been obtained only from CH_3OH or its isotopic species. Recently by means of a special design of the resonator several laser lines around 35 μm were reported in normal CH_3OH (6,7).

The spectral coverage is impressively broad considering that lines up to 1200 μm are also obtained from normal methyl alcohol.

Direct frequency measurements were reported for about 200 lines, contributing to making this laser a unique and efficient source with a wide application in laser frequency synthesis, laser magnetic resonance spectroscopy and spectroscopy of the active medium itself.

In the present work we have pumped for the first time the isotopic species $^{13}CD_3OH$ searching for the generation of new strong emissions, hence further increasing the abundance of lines available from the methanol laser. We report 36 new cw laser lines, many of them at wavelengths shorter than 100 μm.

Experimental arrangement

The FIR resonator, described elsewhere (8), consisted of a 1 m long open structure resonator. The CO_2 pump power in general was coupled into the resonator through a 1 mm hole

in one copper end mirror (2 m radius). A transverse pumping scheme could also be used for the strong lines. In this case the amplitude stability of the system improved marcately due to the absence of 10 μm radiation back reflected into the CO_2 laser. FIR power was coupled out using a variable coupler consisting of an elliptical mirror formed by cutting and polishing a 6 mm diameter copper cylinder at 45°. The mirror was moved perpendicular to the cavity axis to optimize the output coupling at each wavelength. The output radiation was transmitted through a polyethylene window in the side of the vacuum chamber of the laser. A flat gold-coated pyrex mirror at the other end was adjusted by a calibrated micrometer to change the resonant frequency of the cavity. By scanning the cavity length accurate wavelength measurements (with fractional uncertainties between 10^{-3} and 10^{-4}) could be made easily since higher order transverse modes could be controlled by irises at each end of the FIR resonator.

As a pump source a 1 m long high pressure CW quartz waveguide CO_2 laser was used. More than 30 watts were available on the strongest lines and about 10 watts on the weakest ones. For some of the new emissions, direct frequency measurements were performed with the technique, introduced in (9), of synthesizing an appropriate local oscillator signal from the difference frequency of two saturated absorption fluorescence stabilized CO_2 lasers. A conical W-Ni metal-insulator-metal point contact diode was used as mixer (10) and the laser beams were focused on the diode by parabolic mirrors. Pump offset were determined by heterodyning the pumping radiation with an actively stabilized CO_2 laser.

Results and discussion

The new FIR laser emissions, ordered by pump line, are summarized in Table I. The values of pressure, measured with a thermocouple gauge calibrated with a capacitance manometer, are indicative for optimum output. The laser lines whose frequency has been measured are listed in Table II, ordered by increasing frequency.

Confirming the efficiency of methyl alcohol as FIR active medium, many of the observed emissions are "strong". More precisely, power levels higher than 1 mw are obtained, even though the FIR resonator was designed for clean mode structure operation rather than for high power.

We performed a quantitative analysis of the efficiency on the strongest new emission at 127.0 μm. The optimized FIR output power was measured by means of the absolute power meter described in (11). The data as a function of the 10P8 pump power available are reported in Fig.1. For a comparison, also the results obtained with

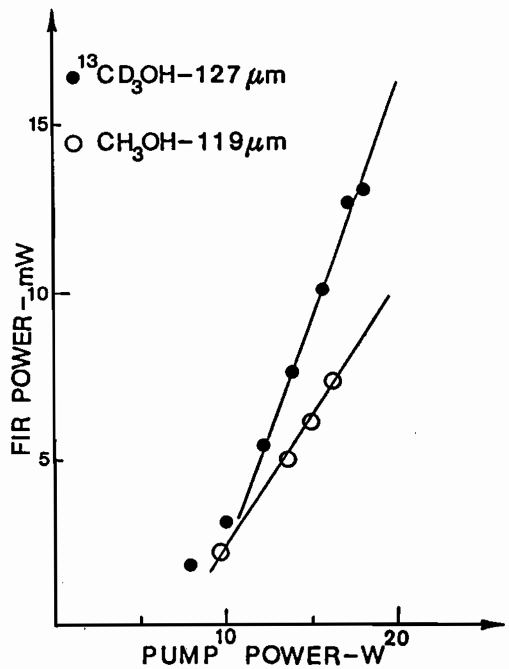

Fig. 1: Measured efficiency for the new laser line at 127.02 μm

the same apparatus on the CH_3OH well known line at 119 μm (9P36 CO_2 pump) are shown. The higher efficiency measured at 127.0 μm suggests that $^{13}CD_3OH$ could become competitive with normal CH_3OH in applications, like plasma diagnostics, where high power is required.

We think that the new discovered lines can expedite spectroscopic investigation in regions were no alternative sources to FIR lasers are available. For instance the new line at 127.6 μm (2348438.4 MHz) has already been applied to the laser Magnetic Resonance detection of atomic Silicon (12).

As a final remark, the present investigation should be considered not complete. We think that the results in the present paper demonstrate that $^{13}CD_3OH$ is an efficient source in the far infrared and even more strong lines can be generated after a more systematic investigation.

TABLE I

CW FAR INFRARED LASER EMISSION FROM OPTICALLY PUMPED $^{13}CD_3OH$

PUMP LINE	FIR WAVELENGTH (μm) (a,d)	REL.POL.	PRESS PASCAL(c)	I
9R40	177.6	//	24	W
9R38(b)	387.2	//	19	MW
	151.	//	27	W
9R34	98.5	⊥	24	MW
9R32	55.8	//	27	M
	52.1	⊥	27	M
9R30	150.2	//	30	VS
9R28(b)	153.69	//	27	S
	336.5	//	27	W
9R18	52.2	//	13.5	M
9R14	119.4		20	S
9R10	65.4	//	16	M

Table I - Continued

PUMP LINE	FIR WAVELENGTH	REL.POL.	PRESS	I
9R8	389.6	//	16	M
	221	//	13.5	MS
	196.2	//	12	WM
9P10	126.1	//	26	S
	72.9	//	23	VS
9P34	399.8	//	13.5	M
10R26	146.32	⊥		M
	197.04	//		VS
	468.96			M
	209.0	//		MW
10R24	145.56	⊥		M
10R22	84.41	⊥	27	S
	127.66	⊥	27	W
	110.0	//	34	W
10R20	73.47	⊥	24	M
10R12	67.8			M
10P8	127.02	//	31	VVS
	462.8			
10P16	333.26	//	16	VS
	340.62	//	16	S
10P22	124.3	//		M
	291.0	//		S
10P42	119.1	//	19	S
	148.3	⊥		S

(a) Wavelength accuracy is from 10^{-3} to 10^{-4}.
(b) Evidence for two different pump offsets.
(c) 133.3 Pa = 1Torr.
(d) Two decimal figures are reported for lines whose frequency has been directly measured (see Tab. II).

Table II

Frequency measurements for 11 cw FAR infrared lasing transitions of $^{13}CD_3OH$ pumped by a CO_2 laser

$^{13}CD_3OH$ LASER LINE λ(μm)	MEASURED FREQUENCY (MHz) (uncertainty:$5x10^{-7}$)	VACUUM WAVENUMBER[a] (cm^{-1})	PUMP LINE	CO_2 FREQ. OFFSET (UNCERTAINTY ± 3MHz)
73.47	4080637.2	136.115405	10R20	+16
84.41	3551805.8	118.475490	10R22	- 8
127.02	2360174.8	78.726957	10P8	0
127.66	2348438.4	78.335473	10R22	+14
145.56	2059531.6	68.698578	10R24	+12
146.32	2048803.6	68.340732	10326	-12
153.69	1950581.6	65.064398	9R28	+ 2
197.04	1521430.9	50.749473	10R26	-30
333.36	899571.7	30.006484	10P16	-15
340.62	880120.4	29.357655	10P16	-17
468.96	639264.6	21.323570	10R26	-20

[a]Calculated from the measured frequency c = 299792458 m/s

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