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$^{12}CH_2F_2$ AND $^{13}CH_2F_2$FAR-INFRARED LASERS: NEW LINES AND FREQUENCY MEASUREMENTS

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ABSTRACT

We report here the discovery of 13 new far-infrared laser lines from $^{12}CH_2F_2$ and seven new lines from $^{13}CH_2F_2$. Most of the new lines were pumped by high-J lines of the 9R branch of a cw-CO_2 laser. Wavelengths range from 97.6 to 616.18 μm. Frequency, pump offset, relative polarization, and relative intensity were measured for most of the new lines.

Key Words: far-infrared laser, difluoromethane, $^{12}CH_2F_2$, $^{13}CH_2F_2$, frequency measurement

1-INTRODUCTION

Difluoromethane ($^{12}CH_2F_2$) together with its ^{13}C isotopomer is a well-known, efficient laser medium for the generation of strong far-infrared laser lines, when optically pumped by CO_2 lasers[1]. Both isotopomers have produced nearly 200 far-infrared (FIR) laser lines with wavelengths in the range

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between 95.6 and 1448.1 μm. This is mainly due to the following conditions: (a) the molecule has a large permanent electric dipole moment (μ=1.9785 D, for $^{12}CH_2F_2$),[2] assuring strong FIR laser transitions, (b) it's intense rotational-vibrational spectrum coincides with the emission bands of CO_2 lasers. The vibrational mode of $^{12}CH_2F_2$ corresponding to the CF_2 asymmetric stretch is centered at 1090.1 cm^{-1} and overlaps with the 9 μm branches of the CO_2 laser.[2,3] The substitution of ^{13}C for ^{12}C produces a small shift of the absorption bands of $^{13}CH_2F_2$ which still fall into the frequency range of the CO_2 laser allowing for the pumping of many far-infrared laser lines.[4] Due to selection rules, most coincidences between the CO_2 laser lines and the absorption lines of the molecules will produce two FIR laser emissions, one polarized parallel and the other perpendicular to the CO_2 pump. Together, $^{12}CH_2F_2$ and $^{13}CH_2F_2$ have produced nearly 200 laser lines with wavelengths in the range between 95.6 and 1448.1 μm,[1] with most in the range 200 to 300 μm. Frequencies of approximately 150 of those lines were measured. The reported laser lines were pumped by cw $^{12}C^{16}O_2$ and $^{12}C^{18}O_2$ lasers.

We report in this work new FIR laser lines, 13 from $^{12}CH_2F_2$ and seven from $^{13}CH_2F_2$. The molecules were optically pumped by a cw CO_2 laser with a high-Q cavity. The laser oscillated on regular-band, sequence-band, and hot-band lines providing for new pumping line-sources.

2-EXPERIMENTAL

The cw CO_2 pump laser is a 1.5 m long Fabry-Perot resonator with a high resolution 163 line/mm grating. The grating allows the operation of the laser in regular-band, sequence-band, and hot-band lines in the 9 μm branch.[5] Regular and nonregular lines reach powers of 30 and 10 W.

The FIR laser cavity has a metal-dielectric rectangular waveguide described in detail elsewhere.[6] The lowest loss modes are those having the electric field polarized parallel to the metal walls, and the attenuation of the FIR laser lines is greater when the electric field is polarized perpendicular to them.[7]

A measurement of the FIR wavelength was determined by varying the cavity length over about ten wavelengths by translating one of the mirrors and measuring the length difference with a micrometer. The wavelength thus

obtained can be accurate to about 0.05 μm, and the corresponding estimated frequency is used to select two CO_2 laser lines for the heterodyne frequency measurement.

A metal-insulator-metal (MIM) diode is used to detect the laser radiation, and also as a mixer for the frequency measurements. To measure the frequency of the FIR radiation it is mixed in the MIM diode with selected radiation from two frequency stabilized CO_2 lasers and from a microwave source.[8] A beat note is generated in the diode, and the FIR frequency is obtained from the equation

$$\nu_{FIR} = n|\nu_1 - \nu_2| \pm m\nu_{\mu wave} \pm \nu_{beat}, \quad (1)$$

where ν_1 and ν_2 are the stabilized CO_2 frequencies, $\nu_{\mu wave}$ is the microwave frequency, ν_{beat} is the beat frequency, and n and m are the order of the harmonics generated by the MIM diode. The same diode is used to measure the offsets of the FIR laser lines. In this case, a frequency-stabilized CO_2 laser set to the same laser line as the pump is mixed in the diode with the pump frequency. The frequency of a microwave source is added to the mixture whenever the pump line is a sequence-band or a hot-band line.

3-RESULTS AND CONCLUSIONS

Table I lists 13 new FIR laser lines produced by $^{12}CH_2F_2$, and seven new lines produced by $^{13}CH_2F_2$. Most of the newly discovered lines were pumped by regular high-J lines of the 9R branch and five of them were pumped by hot-band lines. Their wavelengths range from 97.6 to 616.18 μm, and are concentrated on the 100 to 300 μm range. The relative polarization, relative intensity and the optimum pressure of these lines are also reported in this table. The reported intensities are proportional to the rectified voltage on the MIM diode. For comparison the relative intensity of the strong 119 μm line of methanol in the same FIR cavity was of the order of 10 mV.

Table II lists the frequency measurements in the range 0.5 to 1.5 THz (616.18 to 194.4 μm), the pump offsets, and the calculated wavelengths and wavenumbers.

In conclusion, we have obtained 20 new FIR laser lines from $^{12}CH_2F_2$

and $^{13}CH_2F_2$, and measured the frequencies for most of them. The lines were all pumped by lines of the 9 μm branch of a CO_2 laser, from the regular-band, sequence-band and hot-band lines.

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Table I - New far-infrared laser lines from $^{12}CH_2F_2$ and $^{13}CH_2F_2$

CO_2 Pump Line	Wavelength (μm)	Pressure Pa(mTorr)	Relative Polarization	Relative Intensity (mV)
$^{12}CH_2F_2$				
9R(60)	576.4	44(330)	//	0.4
R(58)	142.9	20(150)	//	1.0
9R(56)	105.1	44(330)	//	1.0
9R(54)	616.179	13(100)	//	0.1
9R(52)′	97.6	27(200)	//	0.2
9R(52)″	194.418	13(100)	//	0.6
9R(52)″	210.130	24(180)	//	0.2
9R(50)	279.9	13(100)	//	0.02
9R(48)	201.936	20(150)	//	4.0
9SR(05)	185.0	13(100)	⊥	2.0
9R(02)	194.484	44(320)	//	1.0
	346.8	33(250)	⊥	2.0
9HP(16)	225.9	27(200)	//	1.0
$^{13}CH_2F_2$				
9R(50)	260.248	27(200)	//	5.0
9R(34)	223.973[c]	40(300)	//	4.0
9R(10)	211.116[c]	27(200)	//	6.0
9R(02)	201.739	33(250)	⊥	0.5

9HP(11)	221.284	13(100)	⊥	5.0
	299.5	11(80)	//	2.5
9HP(12)	195.931	27(200)	⊥	4.0
	271.212	27(200)	//	1.0
9HP(14)	198.548	27(200)	//	1.0
9P(08)	355.126[c]	40(300)	//	0.6

'and " indicate different CO_2 laser frequency offsets.
[c] not a new line. See reference 1.
SR stands for sequence-band line and HP stands for hot-band line.
1 Torr = 133.3 Pa.

Table II - Frequency measurements of laser lines in $^{12}CH_2F_2$ and $^{13}CH_2F_2$

CO_2 Pump Line	Measured Frequency (MHz)	Calculated Wavelength[a] (μm)	Calculated Wavenumber[a] (cm^{-1})	Pump Offset (MHz)
$^{12}CH_2F_2$				
9R(52)"	1 541 998.0	194.418	51.4355	- 17
9R(02)	1 541 479.5	194.484	51.4182	+25
9R(48)	1 484 593.9	201.936	49.5207	- 35
9R(52)"	1 426 697.9	210.130	47.5895	- 17
9R(54)	486 534.6	616.179	16.2290	- 26
$^{13}CH_2F_2$				
9HP(12)	1 530 091.5	195.931	51.0384	- 1
9HP(14)	1 509 921.1	198.548	50.3655	+11
9R(02)	1 486 039.0	201.739	49.5689	+ 2
9R(10)	1 420 034.8	211.116	47.3673[b]	- 46
9HP(11)	1 354 783.8	221.284	45.1907	- 8
9R(34)	1 338 518.1	223.973[c]	44.6482[b]	+26
9R(50)	1 151 949.3	260.248	38.4249	+36
9HP(12)	1 105 380.5	271.212	36.8715	- 1
9HP(11)		299.5[d]		- 8
9P(08)	844 186.3	355.126[c]	28.1590[b]	- 2

[a] calculated from c = 299 792 458 m/s.
[a] estimated uncertainty in the reproducibility of the FIR laser frequency: $\Delta \nu/\nu = 2 \times 10^{-7}$.
[b] calculated from the measured frequency with c= 299 792 458 m/s.
[c] previously reported. See reference 1.
[d] we did not measure the frequency of this line. It has the same offset as the 221.284 μm line.
HP stands for hot-band line.

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