International Journal of Infrared and Millimeter Waves, Vol. 19, No. 5, 1998

NEW FAR-INFRARED LASER LINES AND FREQUENCY MEASUREMENTS OF ACETALDEHYDE AND VINYL FLUORIDE*

E. C. C. Vasconcellos,[1] S. C. Zerbetto,[1] L.-H. Xu,[2] R. M. Lees,[3] L. R. Zink,[4] and K. M. Evenson[4]

[1]*Instituto de Física, Gleb Wataghin*
Departamento de Electrônica Quântica
State University of Campinas, UNICAMP
13083-970 Campinas, SP, Brazil
[2]*Department of Physical Sciences*
University of New Brunswick
Saint John, New Brunswick, Canada E2L 4L5
[3]*Department of Physics*
University of New Brunswick
Fredericton, New Brunswick, Canada E3B 5A3
[4]*Time and Frequency Division*
National Institute of Standards and Technology
Boulder, Colorado 80303-3328

Received March 13, 1998

ABSTRACT

Eight far-infrared laser lines have been obtained by optically pumping acetaldehyde (CH_3CHO) and nine by pumping vinyl fluoride (CH_2CHF) with a cw CO_2 laser. The far-infrared laser structure used a metal-dielectric waveguide cavity. This is the first reported observation of four of the laser lines in acetaldehyde. In this work, we measure the frequency, optimum pressure of operation, relative intensity, relative polarization, and pump offset from CO_2 laser-line center.

Key Words: far-infrared laser, frequency measurement, acetaldehyde (CH_3CHO), vinyl fluoride (CH_2CHF), cw CO_2 laser, new lines.

0195-9271/98/0500-0719$15.00/0

INTRODUCTION

Many lines of optically pumped far-infrared (FIR) lasers have been discovered and measured in different molecules[1] in search of coherent sources to fill some gaps especially in the 30 to 100 μm region, which is very important in ion and molecular spectroscopy.

Acetaldehyde and vinyl fluoride have been investigated before, and the wavelengths of lines have been reported.[1-5] This work presents frequency measurements of new and previously known laser lines which were obtained by optically pumping acetaldehyde (CH_3CHO) and vinyl fluoride (CH_2CHF) with a cw CO_2 laser. Eight far-infrared laser lines have been obtained by optically pumping acetaldehyde (CH_3CHO) and nine by pumping vinyl fluoride (CH_2CHF) with a cw CO_2 laser. The far-infrared laser structure used a metal-dielectric waveguide cavity. This is the first reported observation of four of the laser lines in acetaldehyde. In this work, we measure the frequency, optimum pressure of operation, relative intensity, relative polarization, and pump offset from CO_2 laser line center.

EXPERIMENT

The cw CO_2 laser is a 2 m long, Fabry-Perot resonator with a high-Q cavity.[6] It lases in the regular-band, sequence-band, and hot-band lines with powers up to 40 W in the regular-band lines and up to 15 W in the sequence-band and hot-band lines.

The FIR laser has a 2 m long, metal-dielectric waveguide cavity closed by two, flat end mirrors.[7] It is longitudinally pumped with CO_2 pump radiation entering the cavity through a 1mm diametet hole in one of these end mirrors. A small 45° polished copper mirror near this end couples out a fraction of the FIR radiation. The other end mirror is mounted on a micrometer to tune the cavity into resonance with the FIR radiation. A Brewster-angled Si output window transmits most of the FIR, but blocks most of any residual CO_2 pump radiation. The FIR radiation is detected with either a pyroelectric detector or a metal-insulator-metal (MIM) diode.

MEASUREMENTS

A measurement of the FIR wavelength was carried out by varying the cavity length over about ten wavelengths. This was accomplished by translating one of the mirrors and measuring the length difference with a micrometer. The value 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.[8]

A MIM diode is used as a detector of the laser radiation and also as a mixer for the frequency measurements. The FIR radiation is mixed in the MIM diode with selected radiation from two frequency-stabilized CO_2 lasers and with radiation from a microwave source.[8] The diode generates frequencies of various mixing orders among these four radiation sources. The RF beat note generated in the diode (between 0 and 1.5 GHz) is amplified and observed with a spectrum analyser. Tuning the FIR laser across its gain curve maps out the change in amplitude of the beat note frequency by using a peak-hold feature on the spectrum analyzer. The center of this recorded mapping is then measured with a marker frequency. The FIR frequency is obtained by the equation

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

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 (between 0 and 1.5 GHz which is the bandwidth of our amplifier and spectrum analyzer), and n and m are the order of the harmonics generated by the MIM diode. Observing the beat note display on the spectrum analyzer as the FIR and microwave radiations are changed (one at a time) gives the value and sign of n and m. The same diode is used to measure the offsets of the pump laser (the difference in frequency between the absorption frequency and the CO_2 frequency at line center). 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 radiation and the generated beat note in the diode is the pump offset frequency.

The CO_2 reference lasers are frequency-stabilized to ±10 kHz and their center frequencies are known to an accuracy of 2.5 kHz . The microwave source is accurate within 10 Hz. The largest uncertainty comes from setting the FIR laser to the center of its gain curve. We generally measure each frequency five times or more and report the average of these measurements, and the fractional frequency uncertainty is typically $\approx 2 \times 10^{-7}$ (1σ).

RESULTS

Tables 1 and 2 present the pump line, the wavelength, the relative intensity, the relative polarization, the optimum pressure of operation, and the reference for each observed laser line in acetaldehyde and vinyl fluoride, respectively. The data are presented in order of pump line.

Eight lines with wavelengths in the range 319.154 to 528.277 μm were observed in acetaldehyde; four are new and four others have been reported previously.[1,2] The wavelengths reported previously were not measured with high precision. When the reported values were within 1 to 2.5 μm from our values, we considered the laser line as being reported previously; otherwise we called it new. This is the case for the line 335.282 μm pumped by 9R(30) in CH_3CHO; previous work[1,2] report a line pumped by the same pump line at 343 μm and so we considered the line 335.282 μm new. The wavelengths of all nine lines observed in vinyl fluoride, ranging from 148.137 to 671.128 μm have been reported before[1,3-5].

In this work we also measured the frequency, the relative polarization, and the pump offset from CO_2 laser line center for all observed lines. These data are presented in Tables 3 and 4 together with the calculated wavelength, the calculated wavenumber, and the reference to previous reports for each line of acetaldehyde and vinyl fluoride, respectively. The laser lines are presented in order of increasing wavelength in these tables.

ACKNOWLEDGMENTS

We are pleased to acknowledge financial support for this research from the Conselho Nacional de Desenvolvimento Cientifico e Tecnológico (CNPq-Brazilian government), Fundação de Amparo à Pesquisa do Estado de São Paulo (FAPESP-Brazilian government), NASA grant W-19,167 and the National Institute of Standards and Technology (NIST-US government).

Table 1. Observed laser lines of acetaldehyde (CH_3CHO) by pump line.

CO_2 Pump Line	Wavelength [a] μm	Relat. Intens. μV	Relat. Polar.	Pressure Pa(mTorr)	Ref.
9R(50)	528.277	100	//	8(60)	new
9R(50)	470.967	10	//	11(80)	new
9R(40)	415.356	200	//	20(150)	2
9R(38)	319.154	120	//	8(60)	new
9R(36)	509.706	150	//	20(150)	2
9R(30)	385.102	300	//	13(100)	2
9R(30)	335.282	40	//	13(100)	new
9R(22)	328.784	250	//	40(300)	2

[a] calculated from the measured frequency with c = 299 792 458 m/s.
// stands for parallel.

Table 2. Observed laser lines of vinyl fluoride (CH_2CHF) by pump line.

CO_2 Pump Line	Wavelength [a] μm	Relat. Intens. mV	Relat. Polar.	Pressure Pa(mTorr)	Ref.
10R(50)	148.137	1.0	//	19(140)	3
10R(50)	445.831	0.8	//	19(140)	3
10R(20)	444.797	2.0	//	31(230)	4
10P(6)	332.363	5.0	//	60(450)	5
10P(14)	376.824	0.7	⊥	21(160)	4
10P(36)	671.128	0.6	⊥	11(80)	4
10P(38)	337.095	0.7	⊥	13(100)	5

CO_2 Pump Line	Wavelength [a] µm	Relat. Intens. mV	Relat. Polar.	Pressure Pa(mTorr)	Ref.
10P(48)	275.839	1.5	//	13(100)	3
10P(54)	194.255	2.0	//	13(100)	3

[a] calculated from the measured frequency with c = 299 792 458 m/s.
// and ⊥ stand for parallel and perpendicular.

Table 3. Frequency Measurements of acetaldehyde (CH_3CHO) by increasing wavelength.

CO_2 Pump Line	Calculated [a] Wavelength µm	Calculated [a] Wavenumber cm^{-1}	Measured [b] Frequency MHz	Rel. Pol.	Pump Offset MHz	Ref.
9R(38)	319.154	31.3328	939 334.4	//	- 31	new
9R(22)	328.784	30.4151	911 823.0	//	+6	2
9R(30)	335.282	29.8256	894 150.0	//	+26	new
9R(30)	385.102	25.9671	778 474.6	//	+ 26	2
9R(40)	415.356	24.0757	721 771.9	//	+16	2
9R(50)	470.967	21.2329	636 546.2	//	+10	new
9R(36)	509.706	19.6191	588 167.0	//	+9	2
9R(50)	528.277	18.9295	567 490.8	//	+10	new

[a] calculated from the measured frequency with c = 299 792 458 m/s.
[b] 1σ uncertainty, $\Delta\nu/\nu$ is ≈ 2×10^{-7}.
// stands for parallel.

Table 4. Frequency Measurements of vinyl fluoride (CH_2CHF) by increasing wavelength.

CO_2 Pump Line	Calculated [a] Wavelength μm	Calculated [a] Wavenumber cm^{-1}	Measured [b] Frequency MHz	Rel. Pol.	Pump Offset MHz	Ref.
10R(50)	148.137	67.5051	2 023 751.3	//	-24	3
10P(54)	194.255	51.4788	1 543 295.5	//		3
10P(48)	275.839	36.2531	1 086 840.1	//	+37	3
10P(6)	332.363	30.0875	902 001.9	//	+22	5
10P(38)	337.095	29.6652	889 341.2	⊥	+5	5
10P(14)	376.824	26.5376	795 577.0	⊥	+33	4
10R(20)	444.797	22.4822	673 998.0	//	+3	4
10R(50)	445.831	22.4300	672 435.5	//	-24	3
10P(36)	671.128	14.9003	446 699.1	⊥		4

[a] calculated from the measured frequency with c = 299 792 458 m/s.
[b] 1σ uncertainty, $\Delta\nu/\nu$ is ≈ 2×10^{-7}.
// and ⊥ stand for parallel and perpendicular.

REFERENCES

- • Work of the US-Government, not subject to copyright.

1. N.G. Douglas, Millimeter and Submillimeter Wavelength Lasers, Springer-Verlag, New York, pp. 191 and pp. 200 (1989).
2. B.M. Landsberg, M.S. Shafik, R.B. Butcher, *IEEE J. Quantum Electron.* **QE-17**, pp. 828-829 (1981).
3. M. Redon, C. Gastaud, M. Fourrier, *Opt. Lett.* **9**, pp. 71-72 (1984).
4. A. R. Calloway, E.J. Danielewicz, *Int. J. IR and MM Waves* **2**, pp. 933-942 (1981).
5. F. Temps, H.Gg. Wagner, *Appl. Phys.* **B29**, pp. 13-14 (1982).
6. K.M. Evenson, C.C.-Chou, B.W. Bach, K.G. Bach, *IEEE J. Quantum Electron.* **QE-30**, pp. 1187-1188 (1994).

7. M. Inguscio, F. Strumia, K.M. Evenson, D.A. Jennings, A. Scalabrin, S.R. Stein, *Opt. Lett.* **4**, pp. 9-11 (1979).

8. F.R. Petersen, K.M. Evenson, D.A. Jennings, J.S. Wells, K. Goto, J.J. Jimenez, *IEEE J. Quantum Electron.*, **QE-11**, pp. 838-843 (1975).