OPTICALLY-PUMPED FAR-INFRARED LASER LINES IN HYDRAZINE, METHANOL, HEAVY WATER, AND AMMONIA: NEW LASER LINES AND FREQUENCY MEASUREMENTS

E. C. C. Vasconcellos,[1] M. D. Allen,[2] L. R. Zink,[2] and K. M. Evenson[2]

[1]*Instituto de Física, Gleb Wataghin*
Departamento de Eletrônica Quântica
Universidade Estadual de Campinas
13083-970 Campinas, São Paulo, Brazil
[2]*Time and Frequency Division*
National Institute of Standards and Technology
Boulder, Colorado, 80303-3328 U.S.A.

Received March 15, 2000

ABSTRACT

A far-infrared laser cavity designed to favor short-wavelength laser lines was used to generate optically-pumped far-infrared laser radiation. New far-infrared laser lines were discovered in hydrazine, heavy water, ammonia, and several short-wavelength lines previously discovered in methanol were observed. Wavelength, frequency, and relative intensity measurements were performed on laser lines in the wavelength range 42.4 to 253.7 μm. Each far-infrared frequency measurement was obtained by mixing the far-infrared radiation with radiation from two reference CO_2 lasers and from a microwave synthesizer in a metal-insulator-metal diode.

The pump laser was a high-Q Fabry Perot resonator oscillating on 275 grating-selected laser lines lincluding regular, sequence, and hot band lines.

Key Words: hydrazine, methanol, heavy water, ammonia, new far-infrared laser lines, optically pumped far-infrared laser, laser frequencies, relative intensity.

INTRODUCTION

We have invented a more powerful far-infrared laser cavity that produces more short-wavelength lines than our previous lasers do. Therefore, we tried this laser on hydrazine, methanol, heavy water, and ammonia which have been studied previously as far-infrared (FIR) laser media.[1-4] In this work we discovered new far-infrared laser lines in hydrazine, heavy water, and ammonia, and observed several short wavelength lines previously discovered in methanol. We measured the wavelength, frequency and relative intensity for each line.

LASER DESCRIPTION

We used a far-infrared cavity consisting of a 1 m long, and nominally 25.4mm inside diameter copper tube closed at the ends by two mirrors with 1.2 m radius of curvature. One of the mirrors was movable longitudinally by a micrometer in order to set the FIR cavity in resonance with the FIR radiation.

The CO_2 pump laser was a 1.5 m long, high-Q Fabry-Perot resonator[5] that in its most recent version uses only one grating to cover the whole emission spectrum. The CO_2 laser grating at one end selects the infrared emission lines and is also used to couple out the laser radiation in zeroth order with 3% coupling. The grating has a better selection than most CO_2 lasers because we have eliminated the wall-bounce mode by placing 5 fixed irises in the laser tube. The pump laser operates on 275 laser lines in regular, sequence and hot band lines with powers up to 40 W on the regular lines.

MEASUREMENTS

To pump the FIR laser medium the CO_2 laser radiation was focused into the FIR cavity from one of its ends so as to be reflected back and forth in a horizontal plane nearly perpendicular to the copper tube.

When searching for new FIR laser lines we scanned the CO_2 laser lines and looked for IR absorption by the FIR medium by monitoring the photoacoustic signal from a microphone mounted inside the FIR cavity. Once an absorption signal was found we looked for FIR emission by lowering the pressure to operation level in the range 7 to 53 Pa. We then adjusted the pressure, pump offset, and output coupling in order to optimize the signal. A scan of the FIR laser modes was recorded as a function of the FIR cavity length, from which a first measurement of the number of lines oscillating at the particular pump line and their approximate wavelengths were obtained. For each FIR line selected we measured the length of the cavity scan responsible for 20 $\lambda/2$ or more recorded FIR laser modes. This gave us a wavelength value accurate to approximately $\pm$ .05 μm.

To measure the frequency of the FIR line we mixed its radiation with the radiation of two reference CO_2 lasers and of a microwave synthesizer in a MIM (metal-insulator-metal) diode, generating a beat note in the diode.[5,1] The FIR frequency is obtained from the equation:

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

where ν_1 and ν_2 are the stabilized CO_2 frequencies, $\nu_{\mu wave}$ is the microwave frequency, ν_{beat} is the rf beat frequency generated in the MIM diode, and n and m are the mixing orders of each stabilized CO_2 laser. Once the beat note is measured and the sign of the mixing components is determined, equation (1) gives the resulting FIR frequency.

RESULTS

Table 1 presents the results obtained for hydrazine (N_2H_4), methanol ($^{12}CH_3OH$ and $^{12}CD_3OH$), heavy water (D_2O), and ammonia (NH_3). Eight new laser lines were discovered and their frequencies were measured. Additionally, frequencies of already reported lines were measured for the first time or remeasured. Optimum pressure for line operation and relative powers were also obtained. The wavelengths were calculated by using c = 299 792 458 m/s (λ was calculated from $\lambda = c/\nu_{FIR}$).

Table 1- Summary of FIR lines pumped by a CO_2 laser in hydrazine, methanol, heavy water and ammonia .

CO_2 laser pump line	Frequency[a]	Calculated[b] Wavelength	Calculated[b] Wavenumber	Pressure	Rel Int.	Ref.
	MHz	μm	cm^{-1}	Pa(mT)		
N_2H_4 :						
9P(35)	2946197.19(59)	101.756	98.2746	25(190)		1
10R(40)[c]	3111748.08(62)	96.342	103.7967	32(240)	S	New
10R(20)[c]	2140871.73(43)	140.033	71.4118	24(180)	W	New
10 R(6)		58.67		36(270)	M	New
10SR(23)	2218652.17(44)	135.124	74.0063	27(200)	W	New
$^{12}CH_3OH$:						
9P(56)[c]	2939964.08(59)	101.971	98.0667	24(180)	W	6
	4413563.12(88)	67.925	147.2206	24(180)		7
	5039174.3(10)	59.492	168.0888	28(210)	W	7,8
$^{12}CD_3OH$:						
9R(34)[c]	5571129.3(11)	53.812	185.8329	20(150)	M	2,3
10R(36)[c]	1181588.64(24)	253.720	39.4136	29(220)	S	2,3
10R(36)[c]	4393866.37(88)	68.230	146.5636	29(220)	S	2,3
10R(34)[c]	7075889.7(14)	42.368	236.0263	17(130)	M	2,3
10R(10)[c]	4223061.41(84)	70.989	140.8662	23(170)	M	2,3
10HR(30)[c]	3674369.79(73)	81.590	122.5638	29(220)	W	9
	2109335.39(42)	142.127	70.3599		W	9

Table 1, continued

CO_2 laser pump line	Frequency [a]	Calculated [b] Wavelength	Calculated [b] Wavenumber	Pressure	Rel. Int	Ref.
	MHz	µm	cm^{-1}	Pa(mT)		
D_2O:						
9R(54)[c]	2740101.96(55)	109.409	91.400	9(65)	W	New
9HP(22)		85.9[d]	224.068			New
	1337952.18(27)	224.068	44.6293	11(80)	W	New
NH_3:						
Non Reg[c]. Line**	5778386.8(12)	51.882	192.7462	33(250)		New

** Non Regular line between 10P(24) and 10P(26)

[a] estimated uncertainty in the reproducibility of the FIR laser frequency $\Delta\nu/\nu = 2\times10^{-7}$.
[b] calculated from the measured frequency with c = 299 792 458 m/s.
[c] frequency measured for the first time.
[d] line discovered in a different FIR cavity.

ACKNOWLEDGMENT

We acknowledge financial support from the Conselho Nacional de Desenvolvimento Científico e Tecnológico (Brazil), the Fundação de Amparo à Pesquisa do Estado de São Paulo (Brazil), the Fundo de Apoio à Pesquisa (FAEP)-UNICAMP, and the National Institute of Standards and Technology (U.S.A.).

REFERENCES

1. E. C. C. Vasconcellos, S. C. Zerbetto, L. R. Zink, and K. M. Evenson, "Hydrazine-an excellent optically pumped far-infrared lasing gas: a review," J. Opt. Soc. Am. **B15**, 1839-1849 (1998).
2. S. C. Zerbetto and E. C. C. Vasconcellos, "Far- Infrared Laser Lines produced by Methanol and its Isotopic Species: A Review", Int. J. IR and MM Waves **15**, 889-933 (1994).
3. D. Pereira, J. C. S. Moraes, E. M. Telles, A. Scalabrin, F. Strumia, a. Moretti, G. Carelli, C. a. Massa, "A review of optically pumped far-infrared laser lines from methanol isotopes," Int. J. IR and MM Waves **15**, 1-44 (1994).
4. N. G. Douglas, "Millimeter and Submillimeter Wavelength Lasers" (Springer-Verlag, New York), 1989.
5. 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. Quantum Electron. **QE-11**, 838-843 (1975).
6. E. C. C. Vasconcellos, S. C. Zerbetto, J. C. Holecek, and K. M. Evenson, "Short-wavelength far-infrared laser cavity yielding new laser lines in methanol," Optics Letters **20**, 1392-1393 (1995).
7. S. C. Zerbetto's doctoral dissertation "Lasers Moleculares na faixa de 0.4 à 11 THz e aplicações à espectroscopia", Instituto de Física "Gleb Wataghin," Departamento de Eletrônica Quântica, Universidade Estadual de Campinas, Dezembro 1995.
8. Li-Hong Xu, Ronald M. Lees, Elza C. C. Vasconcellos, Sandra C. Zerbetto, Lyndon R. Zink, and Kenneth M. Evenson, "Methanol and optically pumped far-infrared laser", IEEE J. of Quantum Electron. **32**, 392-399 (1996).
9. Edjar M. Telles, Lyndon R. Zink, and Kenneth M. Evenson, "New FIR laser lines from CD_3OH," Int. J. IR and MM Waves **19**, 1627-1631 (1998).