New N_2H_4 Far Infrared Laser Lines and Frequencies

E. C. C. Vasconcellos, L. R. Zink, G. P. Galvão, and K. M. Evenson

***Abstract*—Measurements have been made for 32 far infrared laser frequencies of optically pumped hydrazine, 30 of which are new. The wavelengths range from 95 to 528 μm. A pair of frequency-stabilized CO_2 lasers were used as standards for the heterodyne frequency measurements. One of the lines can be used as a local oscillator for astronomical searches for $^{14}N^+$, and other lines can be used to study transitions in metastable Ca.**

I. Introduction

FREQUENCY measurements are reported for 32 N_2H_4 laser lines in the wavelength range 95 to 528 μm. All of these lines, except for two, are newly reported here. This sums to 81 reported N_2H_4 laser lines in the wavelength range 81 to 1007 μm. The frequencies of 64% of these lines have been measured.

The N_2H_4 (hydrazine) molecule is a slightly asymmetric top [1] and has a fairly large electric dipole moment of 1.8 Debye [2], [3]. Also, its infrared spectrum shows strong vibrational absorption bands in the frequency range covered by CO_2 lasers [4]. These attributes make N_2H_4 a good lasing medium for CO_2 laser pumped far-infrared (FIR) lasers. N_2H_4 is an efficient optically pumped FIR laser [5]–[9], with 52 reported laser lines. The frequencies of about 40% of these have been measured.

II. Measurements

The FIR cavity is a nearly confocal, 0.85-m long Fabry–Perot resonator. A conventional CO_2 laser with output power up to 40 W in the strongest lines enters the 5-cm-diameter copper tube at one end at an angle of 75° to the laser axis for nearly transverse pumping (Fig. 1). The FIR power leaves the cavity at the other end and is coupled out from a 12.5-μm-thick polypropylene intracavity beam splitter which is rotated a few degrees from the Brewster's angle for maximum output power.

The laser wavelength was determined by varying the cavity length and measuring the length difference over 10 wavelengths with a micrometer. This yields a wavelength determination accurate to about 0.05 μm. This approximate value is then used to select the CO_2 frequencies for the heterodyne measurement.

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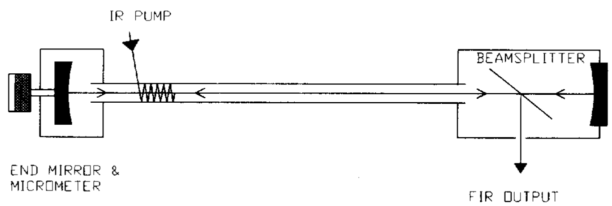

Fig. 1.

The FIR power was detected by a metal-insulator-metal (MIM) point-contact diode which was also used as a mixer for the frequency measurements. The FIR laser frequencies were measured by mixing the FIR radiation with radiation from two frequency stabilized CO_2 lasers and a microwave source on the MIM diode [10]. A beat note

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

is generated between these frequencies, where ν_1 and ν_2 are the stabilized CO_2 frequencies, $\nu_{\mu\text{wave}}$ is the microwave frequency, and

$$O < |\nu_{\text{beat}}| < 1.5\,\text{GHz}. \quad (2)$$

The beat note is amplified and displayed on a spectrum analyzer, using a peak-hold feature that records the beat note as the FIR gain curve is scanned. The center of this recording is then measured using an oscillator, whose frequency is counted.

III. Results

The 32 measured N_2H_4 frequencies are listed in Table I, along with their CO_2 pump lines, optimum pressures, and relative intensities. They range from 0.6 to 3.2 THz (528 to 95 μm), and about 50% of them have wavelengths below 200 μm. Of the 30 newly discovered laser lines, 10 are pumped by 11- μm hot-band laser lines of CO_2. The intensities listed in Table I are proportional to the rectified voltage detected on the MIM diode and should be used as a rough guide to the line's strength. For comparison, the 118.8-μm laser line of CH_3OH had a relative strength of 4200, and at least five strong lines in N_2H_4 showed relative strength in the range 1400 to 4000.

Table II lists all the N_2H_4 FIR laser lines known to date along with their pump laser line, frequency, and reference. Of the 81 listed laser lines, 51 have had their frequencies measured.

IV. Discussion

On each pump line, the CO_2 laser frequency was scanned within its gain profile to maximize the FIR output power.

TABLE I
FAR INFRARED LASER FREQUENCIES OF OPTICALLY PUMPED N_2H_4

CO_2 Pump line	Frequency[a] MHz	Wavelength[b] μm	Pressure[c] Pa(mTorr)	Rel. Intensity
10P(44)	3 172 777.9	94.489	27(200)	800
P(21)HB	2 988 681.2	100.309		
9P(36)′	2 946 196.7	101.756	20(150)	375
P(24)HB	2 823 688.6	106.171	13(100)	450
10R(06)	2 565 990.1	116.833	5(40)	600
10P(44)	2 452 677.4	122.231	27(200)	2500
9P(36)″	2 225 036.9	134.736	27(200)	80
10P(54)	1 980 809.4	151.348	11(80)	
10P(10)	1 880 947.4	159.384	5(40)	80
P(24)HB	1 864 680.0	160.774	10(75)	50
10P(52)	1 860 374.6	161.146		80
P(29)HB	1 858 872.0	161.277	7(50)	1400
10P(52)	1 833 951.7	163.468		
9P(36)‴	1 619 248.8	185.143	13(100)	150
P(20)HB	1 573 986.2	190.467	10(75)	
10P(56)+P(23)HB	1 556 427.9	192.616		4000
10P(24)	1 554 077.8	192.907	15(110)	
P(32)HB	1 282 805.7	233.701	8(60)	1400
10P(40)	1 241 985.4	241.382	7(50)	400
P(32)HB	1 042 133.2	287.672	8(60)	800
P(29)HB	1 040 932.6	288.004	8(60)	500
9P(46)	960 791.1	312.027		
10P(42)	954 857.6	313.966		
	954 850.7	313.968		
9P(12)′	904 899.4	331.299	9(70)	200
9P(12)″	903 889.6	331.669	9(70)	200
10P(56)+P(23)HB	885 606.8	338.516		1500
9R(42)	858 156.2	349.345	5(40)	250
9R(48)	574 117.3	522.180	7(50)	30
10P(42)	569 599.7	526.321		
	569 589.7	526.331		
10P(40)	568 210.2	527.608	7(50)	80

HB indicates 11 μm hot-band line

′, ″, ‴, indicate different CO_2 laser frequency offsets.

TABLE I *(Continued)*

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

TABLE II
SUMMARY OF KNOWN OPTICALLY PUMPED FIR LASER LINES OF N_2H_4

Pump Laser Line	Wavelength N_2H_4[a] μm	Measured Frequency[b] MHz	Reference
CO_2 9R(48)	522.180	574 177.3	NEW
9R(42)	349.345	858 156.2	NEW
9R(22)	327.0		5
9R(18)	368.862	812 750.0	5,8
9R(8)	250.5		5
9P(12)′	331.299	906 899.4	NEW
9P(12)″	331.669	903 889.6*	6
	333.0		5
9P(12)‴	527.873	567 925.4	6
9P(20)	311.075	963 731.4	5,6
	483.5		5
9P(30)	331.5		5
9P(36)′	101.756	2 946 196.7	NEW
9P(36)″	134.736	2 225 036.9	NEW
9P(36)‴	185.143	1 619 248.8	NEW
9P(46)	312.027	906 791.1	NEW
10R(38)	235.56	1 272 681.1	9
	734.162	408 346.7	8
10R(34)	234.0		5
10R(24)	802.4		5
10R(20)	143.246	2 092 854.4	9
	264.801	1 132 140.6	5,6
10R(12)	301.275	995 077.8	5,6
	373.576	802 492.8	5,9
10R(8)	233.916	1 281 625.8	6
	235.0		5
	533.654	561 773.0	9
	533.656	561 771.3	5,8,9

TABLE II *(Continued)*

10R(6)	116.833	2 565 990.1	NEW
10R(4)	150.709	1 989 217.5	9
10P(6)′	134.922	2 221 963.2	9
10P(6)″	137.862	2 174 579.3	9
10P(6)‴	181.926	1 647 877.4	6
	246.5		5
10P(10)	159.384	1 880 947.4	NEW
10P(12)	721.0		5
10P(16)′	81.229	3 690 723.1	9
	102.551	2 923 359.0	9
10P(16)″	461.072	650 207.7	5,6
10P(18)	271.5		5
	372.5		5
10P(22)	1007.0		5
10P(24)	192.907	1 554 077.8*	5,6
	336.0		5
	435.772	687 957.4	5,6
10P(28)	262.0		5
10P(32)	795.0		5
10P(40)	241.382	1 241 985.4	NEW
	527.608	568 210.2	NEW
10P(42)	313.966	954 857.6	NEW
	313.968	954 850.7	NEW
	526.321	569 599.7	NEW
	526.331	569 589.7	NEW
10P(44)	94.489	3 172 777.9	NEW
	122.231	2 452 077.4	NEW
10P(52)	161.146	1 860 374.6	NEW
	163.468	1 833 951.7	NEW
10P(52)+P(23)HB	338.516	885 606.8	NEW
10P(54)	151.348	1 980 809.4	NEW
10P(56)+P(23)HB	192.616	1 556 427.9	NEW
P(20)HB	190.467	1 573 986.2	NEW
P(21)HB	100.309	2 988 681.2	NEW
P(24)HB	106.171	2 823 688.6	NEW
	160.774	1 864 680.0	NEW
P(29)HB	161.277	1 858 872.0	NEW
	288.004	1 040 932.6	NEW

TABLE II *(Continued)*

P(32)HB	233.701	1 282 805.7	NEW
	287.672	1 042 133.2	NEW
$^{13}CO_2$ 10P(18)	219.0		7
10P(24)	945.0		7
10P(30)	289.0		7
$C^{18}O_2$ 10P(24)	705.0		7
$^{13}C^{18}O_2$ 9P(14)	195.0		7
10R(18)	267.0		7
10P(14)	863.0		7
N_2O R(11)	320.0		7
P(7)	200.0		7
P(11)	575.0		7
P(15)	371.0		7
P(24)	237.0		7
P(28)	487.0		7

a Calculated from measured frequency (where applicable), using c= 299 792 458 m/s.
b Accuracy is one part in 10^7.
* Remeasured in the present work.

Different optimum pump frequencies within the same line are denoted by prime, double prime, and so on. We remeasured the frequency of the 192.907-μm line because the reported value in [6] was incorrect and in disagreement with that reported by Douglas [8]. The latter differs from ours by 1.8 MHz, a little over our estimated frequency reproducibility of 2×10^{-7}. We also remeasured the frequency of the 331.669-μm line; the agreement is within the measurement uncertainty.

Several new lines were pumped by 11-μm hot-band laser lines of CO_2 which is a good source for pumping FIR lasers. Two of the FIR lines pumped by 10P(42) are actually sets of doublets, one at $314\,\mu$m and the other at $526\,\mu$m. The doublet frequencies are 6.9 MHz and 10.0 MHz apart. The 10R(8) pump line of CO_2 produces a doublet at $534\,\mu$m, and the doublet frequencies differ by only 1.7 MHz [9]. These doublets preclude using these lines for spectroscopy, such as laser magnetic resonance.

Among the new lines, one is of special interest: the 122.231-μm laser line pumped by the 10P(44) CO_2 laser line is of interest to study the $^{14}N^{+} J = 2 \leftarrow 1$ transition (2 459 380.1 MHz) [11].[1] Carelli *et al.* [13] suggest the CD_3OD (122.30-μm) laser line to study the $^{14}N^{+}\ J = 2 \leftarrow 1$ transition. However, this FIR laser line is pumped by a CO_2 waveguide laser at 55 MHz offset, and hence the N_2H_4 (122.231 μm) laser line, pumped by a conventional CO_2 laser, would be a good alternative. It is 6.7 GHz away from the $^{14}N^{+}\ J = 2 \leftarrow 1$, transition, closer than the CD_3OD laser line, which is 8.2 GHz away. Other lines can be used to study transitions in metastable Ca [14]. The 94.489-μm laser line, also pumped by the 10P(44) CO_2 laser line, is 1.5 GHz away from the Ca $2 \leftarrow 1$ transition (3 174 230.0 MHz). Three other lines: the 190.467-μm laser line, pumped by the P(20) hot-band CO_2 laser line; the 192.616-μm laser line, pumped by the 10P(56) + P(23) hot-band CO_2 laser lines; and the 192.907-μm laser line, pumped by the 10P(24) CO_2 laser line, are 7.2 to 10.4 GHz away from the Ca $1 \leftarrow 0$ transition (1 563 630.0 MHz). The closest line to that transition is the very strong 192.616-μm laser line, with a power level that equals that of the 118.8-μm CH_3OH laser line.

[1] The frequency for this transition reported by Cooksy *et al.* [12] was incorrect.

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