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NEW NH_2H_4 FAR-INFRARED LASER LINES AND THEIR FREQUENCIES[†]

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ABSTRACT

We report 25 new far-infrared laser lines and 26 heterodyne frequency measurements in hydrazine. The frequencies range from 1.0 to 5.5 THz with most of the frequencies between 2.5 and 4.0 THz. The lines were generated in a high frequency, far-infrared Fabry-Perot laser cavity pumped by a CO_2 laser. The cavity has a high Q for wavelengths below 150 μm and uses variable coupling to optimize the power for each line.

INTRODUCTION

Hydrazine (N_2H_4) is one of the most efficient far-infrared (FIR) laser molecules and is now known to also produce short wavelength lines [2] as well as the earlier reported long wavelength lines[1]. Nearly 155 laser lines have been observed by optically pumping hydrazine with CO_2 lasers [1,2]. This work reports 25 new FIR hydrazine laser lines between 1 and 5.5 THz. A low loss, Fabry-Perot FIR laser cavity with variable coupling was used for the FIR laser [3]. Two frequency-stabilized CO_2 lasers served as standards for heterodyne frequency measurements of the 25 new lines, as well as one line reported in [2]. We also report the relative intensity, relative polarization, pump offset, and operational pressure for most of these lines.

EXPERIMENTAL

The pump laser is a 1.5 m long CO_2 laser with a 135 line/mm grating lasing on about 137 different CO_2 lines, including regular and hot band lines. The laser has a ribbed tube that increases the effective resolution of the grating by preventing wall bounces. Both 9 and 10 μm branches operate on high J lines out to 9R(58), 9P(58), 10R(56), and 10P(60); furthermore, 30 hot band lines lase in the 11 μm region. Regular laser lines reach powers up to 25 W and hot band lines up to 6 W.

The FIR Fabry-Perot cavity has less than 0.5% diffraction loss at wavelengths below 150 μm and uses a folded confocal geometry [3]. It consists of a 36 mm diameter, 2 m long precision-bore Pyrex tube closed by a fixed, flat, copper mirror and a 4 m radius-of-curvature gold coated concave mirror. The concave mirror, attached to a micrometer, is movable to tune the cavity into resonance with the FIR laser modes. A 6 mm diameter, 45° copper mirror situated on outer edge of the cavity couples the FIR power out through a window in the opposite side. Moving this mirror in and out of the cavity mode varies the output coupling. The generated FIR radiation exits the cavity through a Brewster angle silicon window and is focused by an off-axis parabolic mirror onto a metal-insulator-metal (MIM) W-Ni diode.

The CO_2 laser radiation is focused into the FIR laser cavity with a 5 m radius-of-curvature concave mirror, 2.5 m from the laser. The pump radiation enters the cavity through a 5 mm diameter hole 15 mm above the center of the fixed, flat mirror. This coupling hole is located outside the FIR mode waist in order to reduce FIR losses. The CO_2 beam hits the center of the curved mirror on the opposite end of the laser. It then reflects back to the flat mirror striking it at the bottom near the wall, where another small mirror reflects the pump beam back to the center of the curved mirror, allowing a second pass of the pump radiation inside the cavity. The polarization of the CO_2 pump radiation can be rotated to observe both perpendicular and parallel FIR laser lines.

Wavelengths were determined by tuning the FIR cavity with the movable laser mirror and measuring the mirror displacement for at least 10 wavelengths of the laser line. This value is accurate within a few parts in 10^3. We measured the far-infrared laser frequencies using the method described in [4] and summarized below.

The radiation from two frequency-stabilized CO_2 lasers were mixed with microwave radiation and the unknown laser radiation on a MIM diode. The unknown frequency υ_{FIR} is calculated by

$$\upsilon_{FIR} = |n_1 \cdot \upsilon_{L1} - n_2 \cdot \upsilon_{L2}| \pm m \cdot \upsilon_{\mu wave} \pm \upsilon_{beat}, \quad (1)$$

where υ_{L1} and υ_{L2} are the saturated-fluorescence stabilized CO_2 laser frequencies, $\upsilon_{\mu wave}$ is the microwave frequency, υ_{beat} is the beat frequency, and the integers n_1, n_2, and m are harmonic numbers. The CO_2 laser frequencies, the microwave frequency, and the harmonic numbers are chosen so that υ_{beat} is less than 1.5 GHz. The harmonic number m is determined by changing $\upsilon_{\mu wave}$ plus one megahertz and observing the change of υ_{beat}. The beat note is amplified and displayed on a spectrum analyzer, using a peak-hold feature that records the signal as the FIR laser is tuned over its gain curve. We measure the center frequency of the recording with a marker frequency. The uncertainty of the CO_2 synthesized frequency is about 15 kHz at all frequencies; the uncertainty in determining the FIR laser is in finding the center of its gain curve. In more than 20 remeasurements we have determined that uncertainty to be $\pm 2 \times 10^{-7}$ for our technique.

We measure the offset by mixing frequency-stabilized CO_2 laser radiation with the pump laser radiation in a MIM diode. For the regular band pump lines, the reference laser operates on the same laser line as the pump. For sequence and hot band pump lines, the reference laser operates on a nearby regular band line and microwave radiation is added to the MIM to give a beat note of about 300 MHz. This beat note is amplified, displayed on a spectrum analyzer, and measured with a frequency marker.

CONCLUSIONS

Table I lists the wavelengths of the new far-infrared laser lines. The polarization relative to the pump laser, operating pressure, pump offset, and intensity are also listed for most of the lines. Sixteen new laser lines have wavelengths shorter than 100 μm. Most of the lines were pumped by lines of the 10P branch of the CO_2 laser. Four of the new laser transitions were pumped by sequence lines, and four others were pumped by hot band lines. The frequency measurements in the range 1.0 to 5.5 THz (289.0 to 54.5 μm) are listed in Table II.

We have increased the number of hydrazine lines to 180. A total of 139 lines are now frequency measured. The great number of strong, high frequency ($\lambda < 100$ μm) laser lines makes optically pumped hydrazine an excellent source for both atomic and molecular spectroscopy.

TABLE I: Far-infrared laser lines from N_2H_4.

CO2 Pump Line	wavelength μm	Offset[b] MHz	Pressure Pa(mTorr)	Rel. Pol.	Rel. Int.[c]
9R(52)	67.159	+43	42(400)	∥	0.3
9R(12)	75.409	+35	59(450)	∥	0.1
9R(10)	73.037	+43	45(350)	∥	0.3
9P(22)	119.419	+39	43(320)	∥	0.4
10P(04)	77.985*	-36	29(220)	∥	5
10P(18)'	64.211	-43	29(220)	∥	0.6
10P(18)"	117.294	+26	27(200)	∥	0.6
10P(20)	84.216	-42	66(500)	∥	3
10P(22)	81.217	+31	20(150)	∥	0.2
	102.558	+31	20(150)	∥	0.4
10P(24)'	81.540	+40	29(220)	∥	7
10P(24)"	121.837	+13	27(200)	∥	2
10P(24)"'	192.9**	-32	27(200)	∥	30
10P(26)	93.761	-46	29(220)	∥	3.5
10P(30)	87.096	-36	13(100)	∥	1
10SP(29)	123.263	-12	27(200)	⊥	50
10P(32)	135.066	0	19(140)	∥	0.2
10SP(31)	63.484	+1	27(200)	∥	2
	84.233	+1	27(200)	∥	6
10SP(33)	54.561	-7	27(200)	∥	1
10P(36)	91.778	-33	21(160)	∥	3
10P(46)	87.029	+34	33(250)	⊥	0.6
10P(54)	100.309		28(210)	⊥	30
10HP(22)	289.046		24(180)	⊥	2
10P(56)+					
10HP(23)	76.938		24(180)	∥	40
10HP(25)	175.455		19(140)	∥	2
10HP(29)	93.264		33(250)	∥	10

* Previously reported [2]. **Previously reported [1].

a. "S" denotes a sequence line, and "H" denotes a hot band line.

b. Estimated uncertainty in the offset measurement is 2 MHz.

c. The 119 μm (9P(36) pumped) CH_3OH line in this laser has a relative intensity of 100. , "and "' denote different offsets.

TABLE II: Far-infrared frequency measurements of optically pumped N_2H_4

λ µm	CO_2 Pump Line	Frequency MHz	Wavenumber cm^{-1}
54.561	10SP(33)	5 494 676.8	183.28269
63.484	10SP(31)	4 722 332.3	157.52005
64.211	10P(18)′	4 668 834.6	155.73556
67.159	9R(52)	4 463 894.4	148.89949
73.037	9R(10)	4 104 665.7	136.91691
75.409	9R(12)	3 975 563.7	132.61053
76.938	10P(56)	3 896 540.2	129.97459
77.985	10P(04)	3 844 230.0	128.22971
81.217	10P(22)	3 691 269.2	123.12749
81.540	10P(24)′	3 676 638.9	122.63947
84.216	10P(20)	3 559 814.8	118.74264
84.233	10SP(31)	3 559 099.1	118.71877
87.029	10P(46)	3 444 748.6	114.90444
87.096	10P(30)	3 442 096.1	114.81597
91.778	10P(36)	3 266 508.2	108.95899
93.264	10HP(29)	3 214 438.5	107.22213
93.761	10P(26)	3 197 414.3	106.65426
100.309	10P(54)	2 988 681.8	99.69169
102.558	10P(22)	2 923 138.5	97.50540
117.294	10P(18)*	2 555 898.1	85.25558
119.419	9P(22)	2 510 428.7	83.73889
121.837	10P(24)*	2 460 606.8	82.07701
123.263	10SP(29)	2 432 134.3	81.12727
135.066	10P(32)	2 219 596.6	74.03777
175.455	10HP(25)	1 708 654.6	56.99458
289.046	10HP(22)	1 037 179.7	34.59659

* Previously reported [2], and now frequency measured.

a. Calculated from the measured frequency with c = 299 792 458 m/s

b. Estimated 1σ uncertainty in the reproducibility of the FIR laser frequency is $\Delta\upsilon/\upsilon=2\times10^{-7}$.

′ and ″ denote different offsets.

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