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Resonant Pumping Far-Infrared NH_3 Laser

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Abstract. A far-infrared (FIR) NH_3 laser was resonantly pumped with a line-tunable infrared (IR) NH_3 laser. The number of the observed FIR laser lines amounted to 33. Most of them belonged to $aR(J,K)$ rotation-inversion transition in $(0,1,0,0)$ vibrational state. The line tunability of sealed-off FIR NH_3 laser was almost achieved in 90, 115, 150, and 220 µm wavelength regions by the selective line tuning of the pumping IR NH_3 laser.

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Far-infrared (FIR) molecular gas lasers pumped with high power infrared (IR) lasers have been developed since 1970 [1], and many laser lines have been obtained. But the line tunability in FIR region was not fully established as compared with that in IR region, for example, CO_2 laser.

In our previous paper [2] it was reported that 24 FIR lines were obtained from NH_3 laser pumped with a TE CO_2 laser. It can be considered that the transitions of most of the FIR laser lines belonged to the off-resonant pumping scheme [3]. The optimum condition of the FIR NH_3 laser, especially the optimum gas pressure, was affected by the deviation of frequency between the pumping CO_2 laser and the corresponding NH_3 absorption line center [4]. This is disadvantageous to make sealed-off lasers that are suited to applications.

In the case of a line-tunable IR NH_3 laser, NH_3 molecules are pumped to $a(6,K)$ sublevels in $(0,1,0,0)$ vibrational state by CO_2 $9R(30)$ radiation and are translated to other rotation-inversion sublevels by fast collisional relaxation. This effect is quickened by buffer gas, for example, N_2 gas [5]. After this relaxation, many vibration-rotation lines have possibility of a laser action. On the other hand, the fast rotation-inversion relaxation is disadvantageous to FIR (rotation-inversion transition) laser action, namely, it will be difficult to obtain a multi-line FIR NH_3 laser action based on collisional relaxation. After consideration of the characteristics mentioned above, we formed a resonantly pumped FIR NH_3 laser.

In Fig. 1, the pumping and emission transitions of "IR NH_3 laser pumped FIR NH_3 laser" are shown schematically. The IR NH_3+N_2 laser is operated as a frequency converter to reduce the deviation of frequency between the pumping and the absorption lines for FIR NH_3 laser. This resonant pumping scheme (cf. [6]) will make it possible to keep the lasing gas pressure of the FIR NH_3 laser constant on every emission line and in addition to obtain some new FIR laser lines. The energy conversion efficiency of this converter is rather low. But it should be emphasized that many FIR laser lines may be obtained by such simple operation as rotation of an IR NH_3 laser grating.

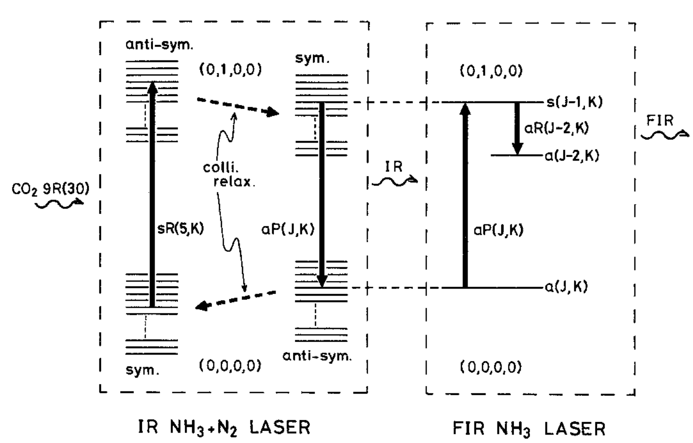

Fig. 1. The schematic transitions of "IR NH_3 laser pumped FIR NH_3 laser"

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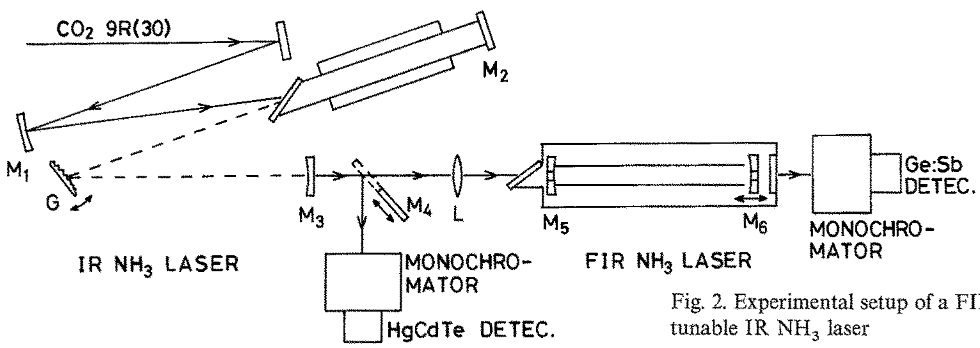

Fig. 2. Experimental setup of a FIR NH_3 laser pumped with a line-tunable IR NH_3 laser

1. Experimental Setup

The experimental setup contained three lasers, i.e. a TEA CO_2 laser (LUMONICS 101), an IR NH_3 laser and a FIR NH_3 laser (Fig. 2).

The TEA CO_2 laser was tuned at $R(30)$ line in 9 μm band and was operated in multi-mode. The pulse energy was 1.5 J with a width of 200 ns (FWHM).

The IR NH_3 laser consisted of two mirrors (M_2 and M_3), one grating (75 lines/mm, $\alpha = 26°45'$) and a commercial rectangular brass tube (14 mm square inner cross section and 1 m length) with a cooling jacket. The optical resonator was formed between a gold coated flat mirror (M_2) and a concave Ge mirror (M_3) with a radius of $R = 10$ m and 80% reflectivity at 10 μm. These two mirrors (M_2 and M_3) are connected with each other through the first order of the rotatable grating (G) with 4 m distance along the optical pass. Lasing gas was a mixture of NH_3 and N_2, and the laser tube was cooled with dry ice.

The FIR NH_3 laser tube was made of a commercial copper tube with 14 mm inner diameter and 90 cm length. Two gold coated concave mirrors (M_5 and M_6) with a radius of $R = 5$ m and 2 mm coupling hole at the center were used. The mirror M_5 was fixed on an end of the laser tube and the mirror M_6 was on a micrometer driven translator at the other end in order that the resonator length could be tuned.

The CO_2 $9R(30)$ radiation was gently focused by the mirror M_1 ($R = 5$ m) into the IR NH_3 laser tube through a ZnSe Brewster window at an off-axis angle of about 5°. The IR NH_3 laser beam, which was essentially separated from the transmitted pumping CO_2 radiation (cf. [7]), was taken out through the mirror M_3 in the wavelength region from 10.7 to 13.3 μm. To monitor the emission wavelength of the IR NH_3 laser, we used a mirror (M_4) in front of the IR NH_3 laser. After a particular line was tuned in by rotation of the grating, the mirror M_4 was removed. This radiation was focused by KRS-5 lens with 20 cm focal length into the FIR NH_3 laser through the KCl Brewster window and the coupling hole of the mirror M_5. The FIR laser output was taken out through the coupling hole of the mirror M_6 and a polyethylene window. The mirror M_6 was tuned at a maximum output point on each emission line, of which the wavelength was monitored by Ge:Sb detector after passing through a monochromator (HITACHI FIS-21). The pulse energy was measured by a pyro-electric detector.

2. Results

The FIR NH_3 laser was successfully pumped with the line-tunable IR NH_3 laser. The number of the observed FIR laser lines amounted to 33. The observed wavelengths are shown in Table 1 with the pumping IR NH_3 laser wavelengths.

Before the measurement of the wavelength, we set the pyro-electric detector in front of the FIR monochromator in order to confirm the presence of FIR radiation. The transmitted pumping radiation was taken away by a quartz filter. The emission wavelength was roughly estimated by the measurement of the output variation with the cavity length tuning.

Pure NH_3 gas (99.9% up) was used as the lasing gas of the FIR laser. The relative output intensity of each line at the optimum pressure is shown in the third column of Table 1. They were decided by a rough estimation with the pyro-electric detector, because the output energy of the lines indicated by "W" was too low to be accurately measured by the pyro-electric detector used in this experiment. The pulse energy of the FIR lines indicated by "S" was about 5 μJ. On the other hand, the pulse energy of each IR laser line was higher than 100 μJ and the most intensive line was 11.711 μm [8] with about 10 mJ. The threshold pumping energy of the FIR laser which depended on the NH_3 gas pressure was measured one by one. For example, it was about 1 mJ/pulse at 2 Torr, and about 0.1 mJ/pulse at 0.4 Torr for 151.6 μm line with the 12.280 μm pumping. The actual incident energy into the FIR cavity was

Table 1. The observed FIR NH_3 laser lines

FIR emission					IR pumping				
Wavelength[a] (observed) [µm]	Frequency (observed) [cm^{-1}]	Relative[b] intensity	Assingment[c] (ν_2)	Frequency[c] (calculated) [cm^{-1}]	Wavelength[d] (observed) [µm]	Frequency (observed) [cm^{-1}]	Relative[b] intensity	Assingment[c] (ν_2 funda-mental)	Frequency[c] (calculated) [cm^{-1}]
59.42	168.3	W	*sR* (6,3)	168.41	12.387	807.3	W	*sP* (8,3)	807.47
67.24	148.7	M	*sR* (5,0)	148.74	12.078	828.0	M	*sP* (7,0)	827.88
76.32	131.0	W	*sR* (4,1)	131.05	11.797	847.7	M	*sP* (6,1)	847.88
76.52	130.7	W	*aR* (7,3)	130.59	13.144	760.8	W	*aP* (9,3)	760.68
79.63	125.6	W	*aR* (7,6)	125.54	13.270	753.6	W	*aP* (9,6)	753.59
87.07	114.9	W	*sR* (3,3)	114.82	11.524	867.5	W	*sP* (5,3)	867.72
88.83	112.6	S	*sR* (3,0)	112.53	11.520	868.1	S	*sP* (5,0)	868.00
90.38	110.6	S	*aR* (6,0)	110.65	12.812	780.5	M	*aP* (8.0)	780.56
90.90	110.0	W	*aR* (6,2)	109.94	12.825	779.7	W	*aP* (8,2)	779.56
91.68	109.1	M	*aR* (6,3)	109.02	12.847	778.4	M	*aP* (8,3)	778.29
92.87	107.7	W	*aR* (6,4)	107.68	12.877	776.6	W	*aP* (8,4)	776.46
94.44	105.9	W	*aR* (6,5)	105.85	12.919	774.1	W	*aP* (8,5)	774.03
96.63	103.5	M	*aR* (6,6)	103.46	12.970	771.0	M	*aP* (8,6)	770.91
106.2	94.16	W	*sR* (2,1)	94.14	11.258	888.3	M	*sP* (4,1)	888.08
112.3	89.05	M	*aR* (5,1)	89.04	12.527	798.3	W	*aP* (7,1)	798.22
113.0	88.50	W	*aR* (5,2)	88.46	12.541	797.4	W	*aP* (7,2)	797.45
114.3	87.49	S	*aR* (5,3)	87.46	12.562	796.1	M	*aP* (7,3)	796.13
116.3	85.98	M	*aR* (5,4)	86.00	12.592	794.2	W	*aP* (7,4)	794.24
119.0	84.03	M	*aR* (5,5)	84.01	12.632	791.6	W	*aP* (7,5)	791.73
133.1[e]	75.13	W	*sR* (1,0)	74.87	11.010	908.3	M	*sP* (3,0)	908.20
147.3	67.89	M	*aR* (4,0)	67.89	12.245	816.7	S	*aP* (6,0)	816.65
147.8	67.66	W	*aR* (4,1)	67.69	12.249	816.4	W	*aP* (6,1)	816.39
149.1	67.07	M	*aR* (4,2)	67.06	12.261	815.6	W	*aP* (6,2)	815.59
151.6	65.96	S	*aR* (4,3)	65.98	12.280	814.3	S	*aP* (6,3)	814.24
155.3	64.39	M	*aR* (4,4)	64.41	12.311	812.3	W	*aP* (6,4)	812.30
215.2	46.47	M	*aR* (3,1)	46.47	11.978	834.9	W	*aP* (5,1)	834.82
218.5	45.77	M	*aR* (3,2)	45.80	11.989	834.1	W	*aP* (5,2)	834.01
223.8	44.68	M	*aR* (3,3)	44.66	12.010	832.6	S	*aP* (5,3)	832.63
296.0	33.78	W	*sQ* (3,1)	33.83	11.258	888.3	M	*sP* (4,1)	888.08
343.3[e]	29.13	W	*sQ* (7,3)	28.64	12.387	807.3	W	*sP* (8,3)	807.47
389.1	25.70	M	*aR* (2,0)	25.68	11.711	853.9	S	*aP* (4,0)	853.82
393.5[e]	25.41	W	*aR* (2,1)	25.45	11.715	853.6	W	*aP* (4,1)	853.55
404.6	24.72	W	*aR* (2,2)	24.74	11.727	852.7	W	*aP* (4,2)	852.72

[a] The accuracy of the wavelength is about $\pm 0.2\%$.
[b] S: strong, M: medium, W: weak.
[c] From [9].
[d] The accuracy of the wavelength is about ± 0.003 µm.
[e] The accuracy of the wavelength is about ± 0.5 µm, because the output was too weak for the wavelength to be measured accurately.

reduced to about one half of the value indicated above by KRS-5 lens and KCl window. Finally, all FIR lines proved to be obtainable at 0.2 Torr NH_3. This phenomenon was somewhat different from the result of our previous experiment [4] with direct CO_2 laser pumping.

All of the observed IR and FIR laser lines were identified to the corresponding transitions in comparison with the Stark spectroscopic results [9], as shown in Table 1. For each J, some $aR(J,K)$ lines with different K value were obtained. So the line tunability of the FIR NH_3 laser was almost achieved in 90 µm ($aR(6,K)$), 115 µm ($aR(5,K)$), 150 µm ($aR(4,K)$), and 220 µm ($aR(3,K)$) wavelength regions. The number of FIR lines with $K=0$ was 6. Some of them were previously observed [2, 10] as "cascade transition line", but we obtained them as "single line" in the present experiment.

3. Discussion

3.1. IR NH_3 *Laser Performance*

Optically pumped NH_3 lasers are usually classified according to pumping geometry. A collinear pumping is characterized by its high conversion efficiency. For a line tunable NH_3 laser, however, the structure becomes complex (cf. [8, 9]), because the output beam should have the same direction for every line and be separated from the background light which consists of the transmitted pumping radiation and amplified spontaneous emissions. On the other hand, this separation will be relatively easily achieved on a zig-zag pumping configuration, which have the disadvantage of the poor conversion efficiency. We adopted the zig-zag pumping configuration in the present experiment. The separation from the background light was achieved and the lasing line was selected in the wavelength region from 10.7 to 13.3 μm by rotation of the grating. Due to the limited resolution of the grating, some of the weak lines indicates by "W" in the eighth column of Table 1 could not be fully separated from the nearby strong lines.

Fry [5] schematically explained why the $aP(J, K)$ transition occupied most of the transitions of the observed laser lines from IR NH_3 laser pumped with CO_2 $9R(30)$ radiation. In addition, Tashiro et al. [8] reported that some $sP(J, K)$ lines were also observable when a line-tunable configuration was adopted to an IR NH_3 laser. In fact, we obtained $32aP(J, K)$ lines, and also obtained $7sP(J, K)$ lines. Some of them [11.258 μm : $sP(4, 1)$, 11.471 μm : $aP(3, 2)$, 11.524 μm : $sP(5, 3)$, 11.797 μm : $sP(6, 1)$, and 12.387 μm : $sP(8, 3)$] could not be obtained in our previous experiment [12] with a multi-line IR NH_3 laser which did not have a structure for line selection.

The optimum NH_3 concentration and the optimum total pressure varied from line to line as we previously reported [12]. In the present experiment, however, two typical mixtures were used. The NH_3 concentration was 3% and the total pressure was 60 Torr for the lines with wavelength shorter than 12.35 μm, and 6% and 20 Torr for the others. It is the aim and end to keep the IR NH_3 laser condition constant all over the tunable range. But we could not realize the sealed-off operation mainly due to the low conversion efficiency of the zig-zag pumping scheme.

3.2. Resonant Pumping

One of the subjects on the optically pumped molecular lasers is how the frequency coincidence between a pumping laser and the absorption line center of lasing gas is achieved. In order to solve this problem, we adopted a essentially resonant pumping scheme, namely "IR NH_3 laser pumped FIR NH_3 laser". It was previously reported [13] that the deviation of frequency between the IR NH_3 laser and the corresponding NH_3 absorption line center was under the influence of the relation between the pumping and the emission transitions, which was schematically devided into "three-level system" and "four-level system". All of the IR NH_3 laser lines used in this experiment excepting three lines (11.520, 11.524, and 12.078 μm) belonged to "four-level system" and were scarcely affected by "ac-Stark effect" [14, 15]. As for the result of the similar measurement to the former [3, 13] about the absorption coefficient of 12.280 μm radiation by NH_3 gas, the frequency deviation was comparable to or less than the Doppler width. So, it was considered that the resonant pumping was almost achieved.

One of the characteristics of the resonant pumping scheme is that the lasing gas pressure is kept constant all over the emission lines, because the pumping radiation is efficiently absorbed by lasing gas even in low pressure region where the width of collision broadening is comparable to the Doppler width. In fact, all the FIR lines inTable 1 were observed even if the FIR NH_3 laser was sealed off at 0.2 Torr NH_3.

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