Two-laser sequential pumping of far-infrared molecular laser†

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## ABSTRACT

We have developed a technique of achieving continuous-wave far-infrared lasing in $NH_3$ using a line-tunable mid-infrared $NH_3$ laser which is optically pumped by a $CO_2$ laser. In the sequential pumping, a number of far-infrared laser lines is obtainable from a single $CO_2$ laser pump line. Continuous-wave laser action on 10 far-infrared lines of $^{15}NH_3$, including 4 new ones, has been observed in the wavelength range from 60 to 400 μm.
**Keywords**: far-infrared laser, mid-infrared laser, optical pumping, ammonia

## 1. INTRODUCTION

Optically pumped molecular lasers are useful sources of coherent continuous-wave (cw) radiation in the far-infrared (FIR) region.[1] They are used for high-resolution laser spectroscopy such as laser magnetic resonance spectroscopy[2] and sideband spectroscopy,[3] plasma diagnostics,[4] secondary frequency standards,[5] and local oscillators for heterodyne detection in radio astronomy and aeronomy.[6] For these applications, it is important to have a large number of available laser lines over a wide frequency region.

In most cases, FIR laser action is obtained on purely rotational transitions in vibrationally excited states. To excite the lasing gas to its upper laser level, an accidental frequency coincidence is needed between its rotation-vibration transition and the pumping radiation of a $CO_2$ or $N_2O$ laser. These accidental coincidences critically limit the number of FIR lines available from conventional optical pumping, especially for light molecules with large line spacings. A histogram showing the number and distribution of optically pumped cw FIR laser lines discovered by 1980 is given in Fig.1.[7] Even though more than 3000 FIR lines have been observed with almost 100 molecules by 1990,[8] the spectral density is still unsatisfactorily low, especially for wavelengths shorter than 150 μm.

We have developed a method, which we call sequential pumping, to produce cw FIR lasing by resonant pumping of the molecular transitions using a line-tunable mid-infrared (MIR) laser[9-15] operating on the same molecule. Our method does not depend on an accidental frequency coincidence in the pumping transition and greatly increases the number of FIR laser lines obtainable from a single $CO_2$ laser line, using a number of MIR laser lines as direct pumping sources.

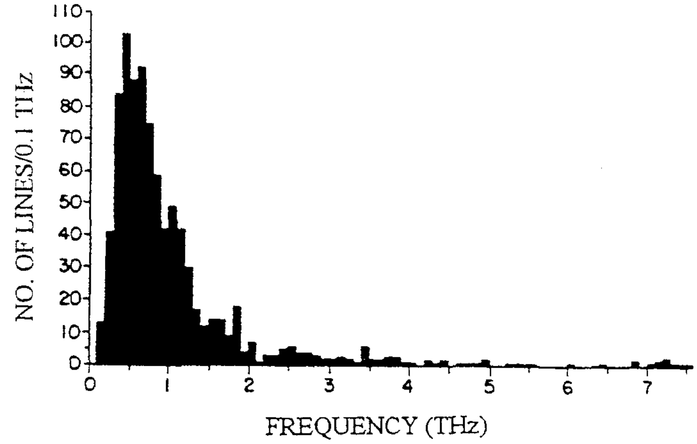


Fig.1 Density of optically pumped FIR laser lines.

## 2. MECHANISM OF SEQUENTIAL PUMPING

The mechanism of the sequential optical pumping of $NH_3$ is shown in the energy-level diagram of Fig.2. In the MIR laser, population is transferred from the ground state ($v_2$=0) to the $v_2$=1 state by optically pumping an R-branch transition with a $CO_2$ laser. Rotational relaxation due to collisions with a buffer gas such as $N_2$ distributes the population from the populated level to its companion rotational levels in the $v_2$=1 state and repopulates the depopulated level by moving population from other rotational levels in the ground state. The population transfer occurs not only within a single *K*-stack but also between different *K*-stacks due to a higher-order collisional interaction. Consequently, population inversion is created on a number of P- and Q-branch lines of the $v_2$ band.

The output radiation from the MIR $NH_3$ laser is used to resonantly pump $NH_3$ molecules to the upper rotational level of a FIR laser transition (see Fig.2) in a separate cavity.[13, 16] Yamabayashi et al.[16] applied the technique to an $NH_3$ laser pumped by a transversely excited atmospheric (TEA) $CO_2$ laser and observed 33 FIR laser lines in pulsed operation. Several groups[13-15] achieved cw oscillation of the MIR $NH_3$ laser, from which an output power of about 1 W is available. We apply the cw MIR laser to what we think is the first sequential optical pumping of a cw FIR laser.

## 3. EXPERIMENTAL SETUP

Figure 3 shows a schematic diagram of the experimental setup. In single $TEM_{00}$ mode operation, the $CO_2$ laser, incorporating a ribbed tube with an active discharge length of 2.4 m and a cavity length of 2.7 m, emits over 55 W power on the 10R(42) line. Detailed design of the $CO_2$ laser is described elsewhere.[17]

The MIR $NH_3$ laser uses a 72 cm long Fabry-Perot cavity, consisting of a copper tube with an inner diameter of 12.7 mm and two end blocks in which the end mirror and the grating are installed. It uses a dichroic ZnSe end mirror that has a radius of curvature of 2 m, transmits 75%

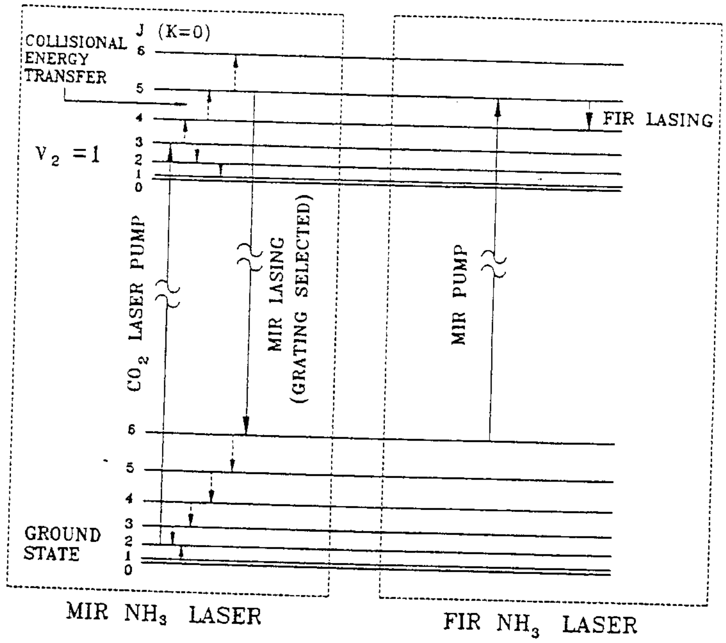


Fig.2 Energy level-diagram showing the pumping mechanism of MIR and FIR $NH_3$ lasers. Only rotational levels in a $K$=0 stack are depicted for simplicity. Collision-induced transitions are indicated by dashed lines.

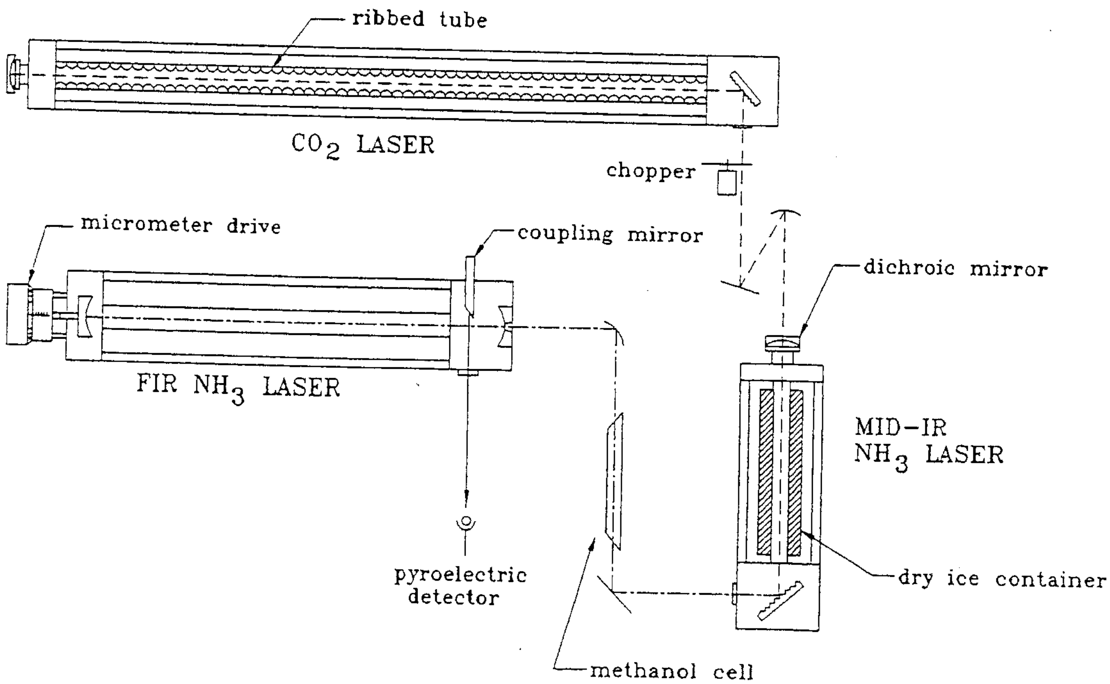


Fig.3 Schematic diagram of the experimental setup.

of the $CO_2$ laser beam, and reflects more than 95% of the $NH_3$ laser radiation. The $CO_2$ radiation is introduced through the end mirror into the laser tube to pump the aR(2,0) transition of $^{15}NH_3$. The pumping beam is slightly focused to achieve better matching with a $TEM_{00}$ mode of the $NH_3$ laser cavity. A gas mixture of 0.3 % $^{15}NH_3$ in $N_2$ at a pressure of 0.43-0.53 kPa (3.2-4.0 Torr) is used. The collisionally broadened $^{15}NH_3$ line overlaps with the pump laser tuned towards the absorption line center even though the frequency offset between the $CO_2$ and $^{15}NH_3$ line centers is 53 MHz.[14] The copper tube is cooled by dry ice to -78 °C, which greatly increases the output power by increasing the population in the lower state of the R-branch transition, and decreasing that in the lower laser level. A 135 line/mm grating with 3 % coupling couples the $^{15}NH_3$ laser radiation out of the cavity in the zero order.

The output beam of the MIR $^{15}NH_3$ laser passes through the $CH_3OH$ cell which absorbs residual $CO_2$ laser radiation. It is then used to pump the FIR $^{15}NH_3$ laser. The 110 cm FIR laser cavity consists of a cylindrical copper tube with an inner diameter of 19.8 mm and two end mirrors. One is gold-coated and the other is made of copper; each has a radius of curvature of 90 cm. The laser appears to be oscillating in waveguide modes for most of the FIR lines obtained here. The gold mirror is attached to a micrometer drive and is movable along the laser axis. The copper mirror has a 1 mm coupling hole at its center and is sealed by a AR-coated ZnSe window. The MIR $^{15}NH_3$ laser beam is introduced into the FIR laser cavity through this hole and resonantly pumps the $^{15}NH_3$ gas along the axis. The inside wall of the laser tube reflects a portion of the diverging pump beam. $^{15}NH_3$ gas flows through the laser tube at a total pressure of 4.0 to 6.0 Pa (30 to 45 mTorr) at the outlet. A fraction of the FIR radiation in the cavity is coupled out of the laser with a 45° copper mirror on a slidable rod inserted perpendicularly into the edge of the cavity mode. A calibrated pyroelectric detector detects the output radiation.

## 4. EXPERIMENTAL RESULTS

The MIR $^{15}NH_3$ laser oscillates on 15 lines in the wavelength range from 11 to 13 μm with a maximum power of 2.0 W (see Fig.4). Replacing the grating by a 10 % coupling one increases the maximum power to 3.4 W, but the number of the lasing lines is reduced to 12. Collisional coupling of ortho-$NH_3$ to para-$NH_3$ is negligibly small at these pressures. Since the directly pumped line is of ortho-$^{15}NH_3$, all the lasing lines are ortho transitions.

With 10 MIR lines as pumping sources, we have observed laser action on 10 FIR lines, as summarized in Table 1 and Fig.4. The wavelength of the laser lines was measured by scanning the FIR modes through several free spectral ranges with the movable mirror. The 1-σ uncertainty of the measurements is estimated to be less than 1 %. Calculation of the wavelength was carried out using the molecular constants in Ref. 18. The observed wavelengths agree with the calculated numbers within the experimental uncertainties, verifying our assignments of the laser lines.

Gastaud et al.[19] previously achieved cw lasing on the sQ(3,3), aR(3,3), sR(4,3), aR(4,3), and aR(5,3) lines by Stark-tuning the molecular transitions into resonance with a $CO_2$ or $N_2O$ laser. Our FIR lines obtained with sequential pumping give slightly different frequencies from those with an applied Stark field. Continuous-wave oscillation on 4 lines (sR(1,0), sR(3,0), sR(5,0), and

Table 1 FIR laser lines observed with optically pumped $^{15}NH_3$. The MIR $^{15}NH_3$ laser was pumped by the $CO_2$ 10R(42) line.

| MIR Laser Lines | Pump Power (mW) | FIR Laser Lines | Wavelength Obs. (μm) | Wavelength Calc. (μm) | Output Power (μW) |
|---|---|---|---|---|---|
| sP(3,0) | 960 | sR(1,0) | 135.5 | 135.97 | 260 |
| sP(4,3) | 1200 | sQ(3,3)[a] | 289.1 | 289.60 | 20 |
| sP(5,0) | 2160 | sR(3,0)[b] | 90.4 | 89.96 | 110 |
| aP(4,0) | 3360 | aR(2,0)[c] | 373.4 | 373.35 | 150 |
| sP(6,3) | 960 | sR(4,3)[a] | 76.4 | 75.98 | 260 |
| aP(5,3) | 1800 | aR(3,3)[a,c] | 220.0 | 218.60 | 450 |
| sP(7,0) | 360 | sR(5,0)[b] | 67.3 | 67.88 | 50 |
| aP(6,0) | 1080 | aR(4,0) | 146.6 | 145.18 | 220 |
| aP(6,3) | 1200 | aR(4,3)[a] | 151.8 | 149.24 | 140 |
| aP(7,3) | 600 | aR(5,3)[a,b] | 113.5 | 113.10 | 130 |

[a] Laser oscillation (cw) was previously obtained by Stark-tuning the molecular transition into resonance with a $CO_2$ or $N_2O$ laser.[19]

[b] Pulsed Raman laser oscillation was previously observed, pumped by a TEA-$CO_2$ laser.[20, 21]

[c] Laser oscillation (cw) was previously observed by conventional pumping with a $CO_2$ laser.[22, 23]

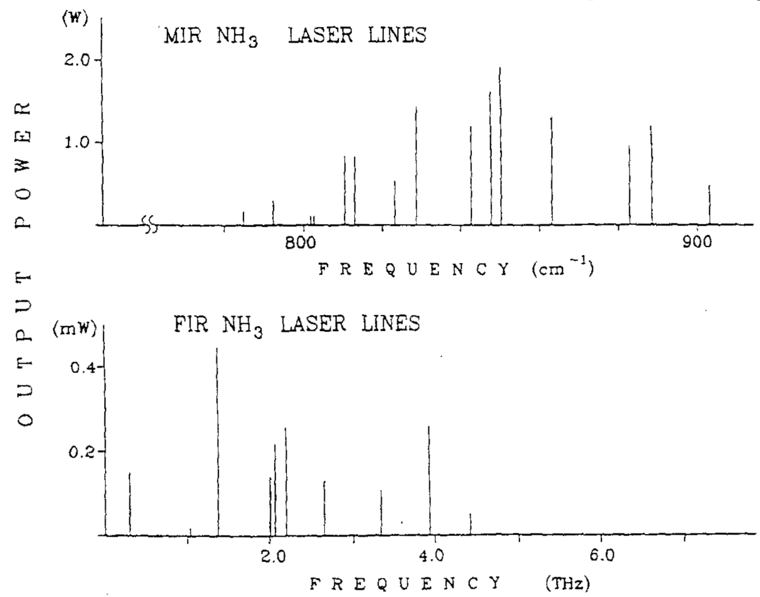


Fig.4 Frequency and output power of the observed MIR and FIR laser lines.

aR(4,0)) has been observed for the first time to our knowledge with this method. We predict that optical pumping of the sP(6,3) line will create population inversion on the inversion transition sQ(5,3). However, the mode competition with higher-order modes on the stronger sR(4,3) line probably prevented the observation of laser oscillation on this line.

## 5. FUTURE PROSPECTS

The sequential pumping is applicable to other MIR $NH_3$ absorptions. More than 60 cw MIR lines have been observed in $^{14}NH_3$; most of these are possible pumping sources for FIR laser lines.[13-15] Ammonia has other close frequency coincidences with isotopic $CO_2$ lasers, sequence- and hot-band $CO_2$ lasers, and $N_2O$ lasers, which have not been tested yet.[22] Use of these pump lasers with a frequency-tunable waveguide cavity will increase the number of MIR lines. Furthermore, it may be possible to achieve MIR laser oscillation from other molecules based on the same mechanism. To create population inversion in the MIR laser, the rotational energy separations in the ground vibrational state must be sufficiently large. Molecules with a relatively large rotational constant $B$, such as deuterated ammonia and water, are possible candidates. FIR lines of these molecules are likely to fall in the wavelength region from 40 to 150 μm, where currently available laser lines are sparse.

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