Sequential optical pumping of a far-infrared ammonia laser

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We present a novel technique for resonantly pumping a continuous-wave far-infrared NH_3 laser with a line-tunable mid-infrared NH_3 laser that is optically pumped by a CO_2 laser. In this two-step process we first convert 10-μm CO_2 laser photons into 11–13-μm NH_3 laser photons, which are then converted into 60–400-μm photons in a far-infrared NH_3 laser. Continuous-wave laser action on 10 far-infrared lines of $^{15}NH_3$, including four new ones, has been obtained with a single CO_2 laser pump line.

Optically pumped molecular lasers[1] are useful sources of coherent continuous-wave (cw) radiation in the far-infrared (FIR) region. They are used for high-resolution laser spectroscopy,[2] plasma diagnostics,[3] and local oscillators for heterodyne detection in radio astronomy and aeronomy.[4]

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 the conventional optical pumping, especially for light molecules with large line spacings. Even though more than 3000 FIR lines have been observed with almost 100 molecules,[5] the spectral density is still unsatisfactorily low, especially for wavelengths shorter than 150 μm.

We present a novel method of using a cw optically pumped line-tunable mid-infrared (MIR) NH_3 laser oscillating in the 11–14-μm wavelength region[6–12] for optical pumping of a separate FIR NH_3 laser, as shown in Fig. 1. A simplified energy-level diagram of NH_3 is shown in Fig. 2. In the MIR laser we transfer population from the ground state ($\nu_2 = 0$) to the $\nu_2 = 1$ state by optically pumping an R-branch transition with a CO_2 laser. The rotational relaxation caused by collisions with a buffer gas such as N_2 distributes the population from the populated level to its companion rotational levels in the $\nu_2 = 1$ 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 because of higher-order collisional interaction. Consequently, population inversion is created on a number of P-branch and Q-branch lines of the ν_2 band.

The output radiation from the MIR NH_3 laser is, of course, resonant with ammonia molecules and can pump them to the upper rotational level of a FIR laser transition (see Fig. 2).[10,13] This is a sequential optical pumping process, and it greatly increases the number of FIR laser lines obtainable from a single CO_2 laser line. Yamabayashi *et al.*[13] applied the technique to a NH_3 laser pumped by a TEA CO_2 laser and observed 33 FIR laser lines in pulsed operation. Several groups of researchers[10–12] achieved cw oscillation of the MIR NH_3 laser, from which a relatively high power near 1 W is available. We applied the cw MIR laser to what we believe is the first sequential optical pumping of a cw FIR laser.

Figure 1 is 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 more than 55 W of output power on the $10R(42)$ line. The design of the CO_2 laser is described in detail elsewhere.[14]

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% of the CO_2 laser beam, and is >95% reflective for the NH_3 laser radiation. The CO_2 laser beam is introduced through the end mirror into the laser tube to pump the $aR(2,0)$ transition of $^{15}NH_3$. There is a frequency offset of 53 MHz between the CO_2 and the $^{15}NH_3$ line centers.[11] The pump beam is slightly focused for better matching with a TEM_{00} mode of the NH_3 laser cavity. A gas mixture of 0.3% $^{15}NH_3$ in 0.43–0.53-kPa (3.2–4.0 Torr) N_2 is used. The collisionally broadened $^{15}NH_3$ line overlaps the pump laser tuned from line center. The copper tube is cooled by dry ice to −78° C, which greatly increases the

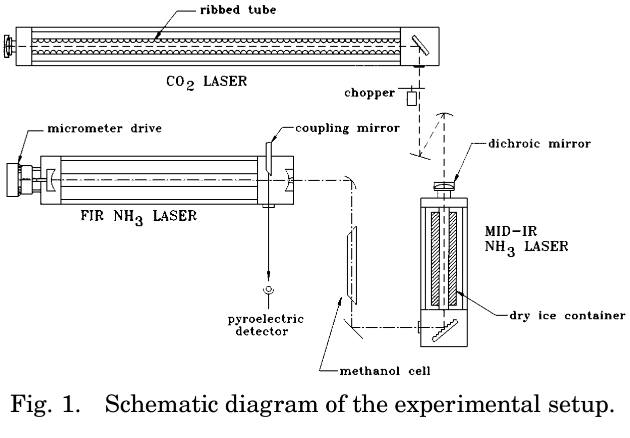

Fig. 1. Schematic diagram of the experimental setup.

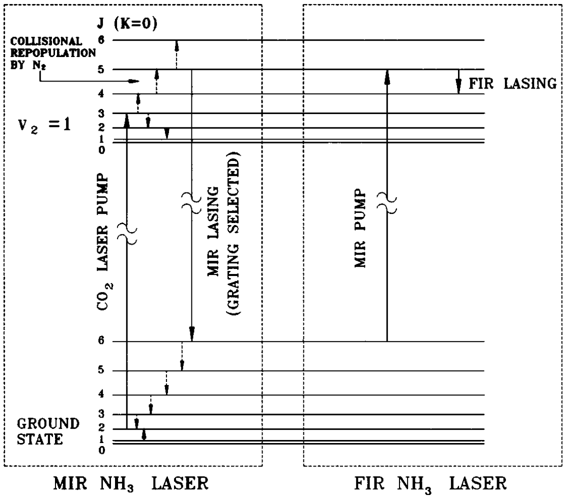

Fig. 2. Energy-level diagram showing the pumping mechanism of MIR and FIR NH_3 lasers. Only rotational levels in a $K = 0$ stack are depicted for simplicity. Collision-induced transitions are indicated by dashed lines. The rotational relaxation caused by N_2 distributes population inversion over a number of MIR lines.

output power by increasing the population in the lower state of the R-branch transition and by decreasing that in the lower laser level. A 135-line/mm grating couples 10% of 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 eliminates 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 mirror is gold coated, and the other is made of copper; each has a radius of curvature of 90 cm. Even though curved mirrors are used, 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 an antireflection-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 4.0–6.0 Pa (30–45 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. The output radiation is detected by a pyroelectric detector calibrated with a powermeter.

The MIR $^{15}NH_3$ laser oscillates on 12 lines with a maximum power of 3.4 W. Collisional coupling of *ortho*-NH_3 to *para*-NH_3 is negligibly small at these pressures. Because 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 observed laser action on 10 FIR lines, as summarized in Table 1. We measured the wavelength of the laser lines 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%. We carried out calculation of the wavelength by using the molecular constants in Ref. 20. The observed wavelengths agree with the calculated numbers within the experimental uncertainties, verifying our assignments of the laser lines.

Gastaud *et al.*[15] previously achieved cw lasing on the same $sQ(3,3)$, $aR(3,3)$, $sR(4,3)$, $aR(4,3)$, and $aR(5,3)$ lines as we observed here by Stark tuning the molecular transitions into resonance with a CO_2 or a N_2O laser. Our FIR lines obtained with sequential pumping give slightly different frequencies from those with an applied Stark field. Cw oscillation on four lines [$sR(1,0)$, $sR(3,0)$, $sR(5,0)$, $aR(4,0)$] was 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.

The sequential pumping is applicable to other MIR NH_3 absorptions. More than 60 cw MIR lines

Table 1. FIR Laser Lines Observed with Optically Pumped $^{15}NH_3$[a]

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

[a] The MIR $^{15}NH_3$ laser was pumped by the CO_2 $10R(42)$ line.
[b] Laser oscillation (cw) was previously obtained by Stark tuning the molecular transition into resonance with a CO_2 or a N_2O laser.[15]
[c] Pulsed Raman laser oscillation was previously observed, pumped by a TEA CO_2 laser.[16,17]
[d] Laser oscillation (cw) was previously observed by conventional pumping with a CO_2 laser[18,19]

have been observed in $^{14}NH_3$; most of these are possible pumping sources for FIR laser lines.[10–12] 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.[18] Use of these pump lasers with a frequency-tunable waveguide cavity will increase the number of MIR lines. Furthermore, there is a possibility of achieving MIR laser oscillation from other molecules based on the same mechanism. For population inversion to occur 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 for lasing gas. FIR lines of these molecules are likely to fall into the wavelength region from 40 to 150 μm, where currently available laser lines are sparse.

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