Far-infrared continuous-wave laser emission from H_2O and from NH_3 optically pumped by a CO laser

M. Schneider* and K. M. Evenson

National Institute of Standards and Technology, Boulder, Colorado 80303-3328

J. W. C. Johns

Steacie Institute of Molecular Sciences, National Research Council, 100 Sussex Drive, Ottawa K1A OR6, Canada

Received March 6, 1996

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We report what we believe is the first observation of continuous-wave far-infrared (FIR) laser emission from water vapor and from ammonia optically pumped by a CO laser. A 2.7-m-long continuous-wave CO laser with 5 W of output power on the stronger lines was used to optically pump either water ($H_2{}^{16}O$) or ammonia ($^{14}NH_3$) in a longitudinally pumped, 2-m-long FIR laser. The FIR laser consisted of a rectangular metal–dielectric waveguide constructed by a pair of horizontal Cu plates separated by a pair of 6-mm-tall glass bars epoxied between the edges of the Cu plates. The glass bars were spaced 50 mm apart and formed a vacuum-tight waveguide with inside dimensions of 6 mm × 50 mm. Flat mirrors were used on each end of the waveguide, and a micrometer was used to tune the laser by moving one of the end mirrors. The dominant mode has its electric field parallel to the copper surface. FIR power was coupled out with a variable coupler consisting of a 5-mm-diameter movable Cu rod, polished at an angle of 45°. It was positioned near one edge of the waveguide and reflected power out perpendicularly to the laser axis, with the beam leaving the cavity on the opposite edge of the waveguide through a Brewster-angle crystal Si window. A Ge bolometer was used to detect the FIR laser radiation.

The CO laser radiation was focused into the cavity through a 1-mm hole in the center of the fixed end mirror with a 1-m-radius concave mirror. Coincidences of H_2O and NH_3 with the CO laser lines were detected with a microphone element placed in a hole in the Cu laser plate near the 1-mm input hole for the CO laser radiation. A 1.5-mm-diameter hole between the microphone and the laser cavity provided a path for the acoustic signal to reach the microphone.

The FIR laser cavity's characteristics can be compared with those of other FIR laser cavities by a comparison of its threshold and the output-power/input-power ratio for the well-known 119-μm line of CH_3OH pumped by the $9P(36)$ line of the CO_2 laser with those of other FIR lasers. The threshold for the 119-μm line was 0.3 W, and the FIR power from this line was 3.5 mW when it was pumped with 4.6 W of CO_2 radiation.

We observed two lasing lines of water at a pressure of 13.3 Pa (100 mTorr) and one line in NH_3 at a pressure of 20 Pa (150 mTorr). The FIR lines had output powers of a few microwatts each. The lines and assignments are shown in Table 1.

We readily assigned the IR H_2O absorptions by comparing the water line frequencies[2,3] with those of the CO laser.[1] The agreement of the pump and absorption lines is within the tunability of the CO laser plus the Doppler width of the H_2O absorption lines. We measured the FIR wavelengths by scanning the FIR modes through some 4–8 free spectral ranges by moving the FIR end mirror. The wavelengths are in agreement with the wavelengths calculated from the frequencies given in Ref. 3.

With NH_3 in the cell and the CO laser oscillating near the $P_9(19)$ line, a strong FIR laser line was observed at 57.4 μm. At first, it was unclear whether the CO laser was oscillating on the $v'' = 10$, $J'' = 13$ line at 1833.526 cm^{-1} or the $v'' = 9$, $J'' = 19$ line at 1834.579 cm^{-1}. However, a Doppler-limited spectrum of NH_3, taken with a Fourier-transform spectrometer at the National Research Council in Ottawa and calibrated with lines of CO given by Brown and Toth,[5] revealed an NH_3 absorption at 1834.57826 cm^{-1}, indicating that the laser was oscillating on the $P_9(19)$ line of CO.

The assignment of the NH_3 absorption line is by no means obvious. According to predicted line positions given by Urban *et al.*,[6] a possible candidate is $s\text{-}R(8, 1)$ at 1834.9730 cm^{-1}. The difference between this prediction and the observed frequency is far greater than the experimental error, but it is possible to calculate a better frequency from the forbidden lines listed more recently by Urban *et al.*[4] The line $a\text{-}P(10, 2)$ at 1472.4593 cm^{-1} has the same upper level as $s\text{-}R(8, 1)$, and, by adding the ground-state energy $a(10, 2)$ as given in Ref. 4 to the forbidden line and subtracting the ground-state energy $s(8, 1)$, we find the frequency of $s\text{-}R(8, 1)$ to be 1834.5786 cm^{-1}, in excellent agreement with the observed frequency.

With the CO laser pumping the level $s(9, 0)$ in ν_4, the lower level of the FIR lasing transition must be $a(8, 0)$. Its energy can be obtained either from the allowed transition $a\text{-}R(7, 1)$ or from the forbidden transition $s\text{-}P(9, 2)$. As before, the frequency predicted from the allowed transition given in Ref. 6 is not close to

0146-9592/96/141038-02$10.00/0

Table 1. Far-Infrared Laser Assignments

CO Laser Pump		Laser Gas Absorption			FIR Laser Emission		
Transition[a]							Calc. λ (μm),
$P_{v''}(J'')$	ν (cm^{-1})	Molecule	Transition	Frequency ν (cm^{-1})	Observed[b] λ (μm)	Transition	Calc. ν (cm^{-1})
$P_7(21)$	1876.63200(1)	H_2O	$\nu_2(7_{34} \leftarrow 7_{07})$	1876.63212[d]	143.3(7)	$(7_{34} \leftarrow 7_{25})$	142.284[c] 70.282
$P_{10}(22)$	1796.92715(1)	H_2O	$\nu_2(8_{54} \leftarrow 8_{45})$	1796.92486[d]	64.9(4)	$(8_{54} \leftarrow 8_{45})$	65.543[c] 154.936
$P_9(19)$	1834.57938(1)	NH_3	$\nu_2 s(9_0 \leftarrow 8_1)$	1834.5783[e]	57.4(4)	$(9_0 \leftarrow 8_0)$	56.860[f] 175.870

[a]Ref. 1 (the tuning range of this CO laser is ± 0.001 cm^{-1}).
[b]Uncertainties are 1 σ estimates.
[c]Ref. 3.
[d]Ref. 2.
[e]This work.
[f]Ref. 4.

an observed line, but a prediction based on the forbidden line listed in Ref. 4 gives 1815.6296 cm^{-1}, in agreement with a line measured at 1915.62597 cm^{-1}. The FIR transition frequency in ν_4 can thus be obtained either from the two allowed lines (combined with appropriate ground-state energies) to give 175.87042 cm^{-1} or from the forbidden lines to give 175.8699 cm^{-1}. These frequencies imply a FIR wavelength of 56.86 μm, which agrees well with the observation at 57.4 μm. Weber[7] did not observe the line at 1834.5876 cm^{-1} in his Stark effect study despite the very good overlap with the $P_9(19)$ CO laser line. He has since informed us that he was unable to get his CO laser to oscillate on this line.

These three FIR lines are at wavelengths where there are not many FIR lasing lines. If a more powerful CO laser is developed, these and other new CO pumped lines may become useful spectroscopic sources.

We thank J. K. G. Watson for his assistance in assigning the NH_3 lines.

*Present address, Forschungsinstitut der Zementindustrie, Düsseldorf, Germany.

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