Far-infrared continuous-wave laser emission from H$_2$O and from NH$_3$ optically pumped by a CO laser

M. Schneider* and K. M. Evenson

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

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We report what we believe is the first observation of continuous-wave far-infrared laser emission from water vapor and from ammonia optically pumped by a CO laser. 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$_2$O absorptions by comparing the water line frequencies with those of the CO laser. The agreement of the pump and absorption lines is within the tunability of the CO laser plus the Doppler width of the H$_2$O 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 $\nu'' = 10$, $J'' = 13$ line at 1833.526 cm$^{-1}$ or the $\nu'' = 9$, $J'' = 19$ 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, 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., a possible candidate is $s$-$R(8,1)$ at 1834.973 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. The line $a$-$P(10,2)$ at 1472.4593 cm$^{-1}$ has the same upper level as $s$-$R(8,1)$, 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 $s$-$R(8,1)$ to be 1834.5786 cm$^{-1}$, in excellent agreement with the observed frequency.

With the CO laser pumping the level $s(9,0)$ in $v_4$, the lower level of the FIR lasing transition must be $a(8,0)$. Its energy can be obtained either from the allowed transition $a$-$R(7,1)$ or from the forbidden transition $s$-$P(9,2)$. As before, the frequency predicted from the allowed transition given in Ref. 6 is not close to...
Table 1. Far-Infrared Laser Assignments

<table>
<thead>
<tr>
<th>CO Laser Pump Transition $^a$</th>
<th>Laser Gas Absorption</th>
<th>FIR Laser Emission</th>
</tr>
</thead>
<tbody>
<tr>
<td>$P_7(21)$ 1876.63200(1)</td>
<td>H$<em>2$O $v_2(7_34 \rightarrow 7</em>{07})$ 1876.63212$^d$</td>
<td>Observed$^b$ $\lambda$ ($\mu$m) Transition</td>
</tr>
<tr>
<td>143.3(7)</td>
<td>(7_{34} \rightarrow 7_{25}) 142.284$^e$ 70.282</td>
<td></td>
</tr>
<tr>
<td>$P_{10}(22)$ 1796.92715(1)</td>
<td>H$<em>2$O $v_2(8</em>{54} \rightarrow 8_{45})$ 1796.92486$^d$</td>
<td>64.9(4) (8_{54} \rightarrow 8_{45}) 65.543$^e$</td>
</tr>
<tr>
<td>154.936</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$P_9(19)$ 1834.57938(1)</td>
<td>NH$_3$ $v_2 s(9_0 \rightarrow 8_1)$ 1834.5783$^o$</td>
<td>57.4(4) (9_0 \rightarrow 8_0) 56.860$^f$</td>
</tr>
<tr>
<td>175.870</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$^a$Ref. 1 (the tuning range of this CO laser is ±0.001 cm$^{-1}$).

$^b$Uncertainties are 1 $\sigma$ 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 $v_4$ 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 $\mu$m, which agrees well with the observation at 57.4 $\mu$m. 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.

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

References


