

# Heterodyne frequency measurements on N<sub>2</sub>O at 5.3 and 9.0 μm

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Heterodyne frequency measurements on the 01<sup>1</sup>1–00<sup>0</sup> band of N<sub>2</sub>O have been made with the use of a tunable-diode laser, a CO laser transfer oscillator, and a CO<sub>2</sub> laser frequency synthesizer. A beat frequency was measured between a CO laser and a tunable-diode laser whose frequency was locked to the peak of N<sub>2</sub>O absorption features. The frequency of the CO laser was simultaneously determined by measuring the beat frequency with respect to a reference synthesized from two CO<sub>2</sub> lasers. New rovibrational constants are given for the 01<sup>1</sup>1 state of N<sub>2</sub>O, which are in excellent agreement with previous results, although the band center is 4 MHz higher than in the previous measurements. A table of the line frequencies and their absolute uncertainties is given for the N<sub>2</sub>O absorption lines in the wave-number region from 1830 to 1920 cm<sup>-1</sup>. Some additional frequency measurements near the lower-frequency end of the 02<sup>0</sup>0–00<sup>0</sup> band have also been made with respect to a <sup>12</sup>C<sup>18</sup>O<sub>2</sub> laser.

## INTRODUCTION

In recent years we have made heterodyne frequency measurements of various molecular absorption features in order to provide benchmark frequency-calibration standards throughout the infrared spectral region.<sup>1–5</sup> Nitrous oxide (N<sub>2</sub>O) is a convenient gas to use for the calibration of spectrometers and certain tunable infrared laser devices because it is a common, stable, easy to handle gas that has strong, sharp, and conveniently spaced absorption features. In this paper we present the results of new frequency measurements made on the 1880-cm<sup>-1</sup> band of N<sub>2</sub>O using some of the same techniques used in the measurements of the 1711-cm<sup>-1</sup> band of OCS (Ref. 2) and the fundamental band of DBr.<sup>3</sup> We also report some measurements on high-*J* transitions in the *P* branch of the 02<sup>0</sup>0–00<sup>0</sup> band and associated hot bands near 1110 cm<sup>-1</sup>.

The best measurements of the wave numbers of the 01<sup>1</sup>1–00<sup>0</sup> band by an alternative technique is the Fourier-transform measurement reported by Amiot and Guelachvili.<sup>6</sup> The intensities of the lines of the 01<sup>1</sup>1–00<sup>0</sup> band were measured by Toth and Farmer.<sup>7</sup>

The wave numbers for the 02<sup>0</sup>0–00<sup>0</sup> band and associated hot bands were given by Olson *et al.*<sup>8</sup> from an analysis of published microwave measurements and infrared hot-band measurements on higher-frequency-band systems. More recently, Guelachvili has made direct measurements on this band using Fourier-transform spectroscopy.<sup>9</sup> Brown and Toth<sup>10</sup> have given even more-accurate measurements. Some of the hot-band transitions measured in the present work were not previously reported.

## EXPERIMENTAL DETAILS

Each of the 5.3-μm measurements consisted of two simultaneous heterodyne frequency measurements. In one mea-

surement, radiation from a tunable-diode laser (TDL) (whose frequency was locked to some N<sub>2</sub>O absorption feature) was heterodyned with CO laser radiation, and the beat-frequency difference was recorded. The second measurement consisted of measuring the CO frequency by comparing its frequency with a reference synthesized from CO<sub>2</sub> laser frequency standards. The N<sub>2</sub>O frequency is then the measured CO frequency algebraically combined with the TDL–CO laser difference-frequency value. Similar measurements have been made on OCS (Ref. 2) and DBr.<sup>3</sup> We describe the procedure briefly and refer to earlier work for additional details. Reference 2 contains a block diagram of the measurement scheme.

The particular TDL mode that was transmitted through the monochromator was divided into two parts. One went to the HgCdTe detector for heterodyning with the CO laser; the other was used for locking the frequency of the TDL to that of some N<sub>2</sub>O line. The use of a first-derivative lock was made possible by adjusting the monochromator to produce a zero-slope background on either side of the N<sub>2</sub>O feature of interest. The frequency-modulation amplitude was made small enough that the jitter linewidth was not increased, subject to the condition that the signal-to-noise ratio (SNR) of the derivative was adequate for locking purposes.

The SNR was high enough for most of the measurements. It was particularly good for the Q-branch measurements, as evidenced by the spectrum shown in Fig. 1. The uncertainty in the TDL–CO laser beat-frequency measurement was taken as one tenth of the jitter linewidth plus one half of the Doppler linewidth (FWHM) divided by the derivative SNR.

The CO laser was used as a transfer laser in these experiments, and we believe that we can determine the value of the CO transition to within 5 MHz. This uncertainty need not apply to the N<sub>2</sub>O frequency determinations, however, since the CO laser frequency is adjusted to a value synthesized from CO<sub>2</sub> laser frequency standards during the N<sub>2</sub>O measurements.

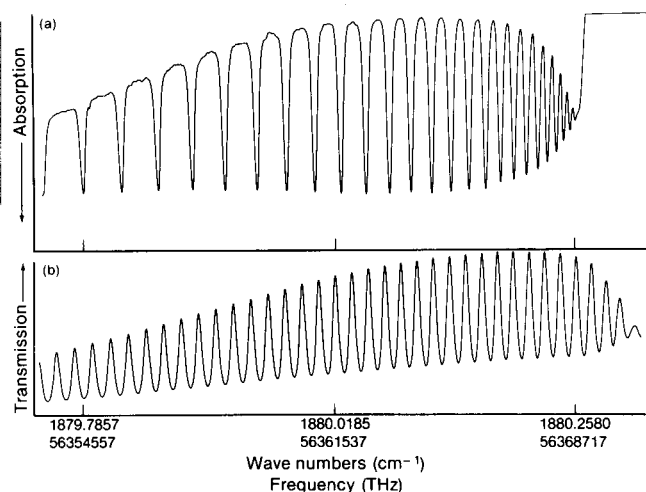


Fig. 1. (a) Portion of the Q branch of the  $01^{11}-00^0_0$  transition in  $N_2O$ . The pressure was 0.53 kPa, and the path length was 1.7 m. (b) Fringe pattern generated by a 76.2-mm-long Ge étalon.

We have adopted this procedure because of observed discrepancies between published CO values and our own measured values. Part of this difficulty may be due to the lack of a widely used stabilization scheme<sup>11</sup> for the CO laser.

The apparatus for synthesizing a reference frequency for the CO laser consisted of two stabilized  $CO_2$  lasers and a metal-insulator-metal (MIM) diode, which served as a harmonic generator and mixer. It was not necessary to use a klystron as a third oscillator. The  $CO_2$  lasers were locked to the center of the saturated absorption dip, which was observed in fluorescence from an intracavity cell.<sup>12</sup> Radiation from the two  $CO_2$  lasers impinged upon the MIM diode; a beat frequency,  $\nu_B$ , propagated from the diode, was amplified and then displayed on a rf spectrum analyzer. The beat frequency was equal to

$$\nu_B = \nu_{CO} - (l\nu_1 + m\nu_2),$$

where  $\nu_1$  and  $\nu_2$  are frequencies of the  $CO_2$  laser standards<sup>13,14</sup> and  $\nu_{CO}$  is the CO laser frequency that is being measured. The harmonic numbers  $l$  and  $m$  are either negative or positive integers. Beat frequencies ranging between 20 MHz and 1 GHz are displayed on the spectrum analyzer and marked with the output of a rf synthesizer. The CO laser is tuned over its gain bandwidth, and the beat frequency undergoes a corresponding excursion on the spectrum analyzer. The center of this excursion is taken as the indication of the center. The CO laser is held at this value for the  $N_2O$  measurement. For the measurements in Table 1 the values for  $l + m$  were either  $1 + 1$  or  $3 - 1$ , and our values for the CO transition are indicated there.

The 9- $\mu m$  measurements were made by heterodyning the frequency-locked TDL with a  $^{12}C^{18}O_2$  laser. In this case the 2-m  $CO_2$  laser was not equipped with an internal stabilization cell, and the center frequency for the transition was determined by tuning the  $^{12}C^{18}O_2$  laser over its gain bandwidth and determining its center from the midpoint of the TDL- $CO_2$  laser beat-frequency excursion. This interpolation was aided by a digital voltmeter on the  $CO_2$  laser's piezoelectric transducer (PZT) driver. Suitable care was taken to avoid possible error resulting from hysteresis in the PZT. This procedure resulted in increasing the uncertainty in the  $N_2O$  measurement because of the uncertainty in finding the center of the  $CO_2$  laser line.

Mesa stripe TDL's were used for both the 5.3- and the 9.0- $\mu m$  measurements. The modes selected for these measurements were those that were both reasonably powerful and had good tuning characteristics in the vicinity of the lines being measured. Figure 1 shows a portion of the  $N_2O$   $01^{11}-00^0_0$  Q branch covered by one mode.

The absorption cell for these measurements was 1.7 m long, and pressures ranged from 0.53 to 2.0 kPa (4 to 15 Torr) for

Table 1. Heterodyne Frequency Measurements on the  $01^{11}-00^0_0$  Band of Nitrous Oxide

Transfer Laser		$N_2O - CO$ Difference Frequency (MHz)	$N_2O$ Transition	Nitrous Oxide	
CO Transition <sup>a</sup>	Measured Frequency (MHz)			Measured Frequency (MHz) <sup>b</sup>	Obs. - Calc. (MHz)
$P_9(19)$	54 999 306.6	+2825.8	$P(50)$	55 002 132.4(50)	-2.2
$P_9(15)$	55 489 266.5	+2459.2	$P(33)$	55 491 725.7(50)	-2.1
$P_8(18)$	55 880 995.3	-5103.8	$P(19)$	55 875 891.5(100)	-6.7
$P_8(15)$	56 250 161.9	-7755.7	$P(5)$	56 242 406.2(50)	1.3
$P_8(14)$	56 371 259.1	-2540.3	$Q(3)$	56 368 718.8(100)	2.6
		-2696.4	$Q(4)$	56 368 562.7(100)	-0.4
		-2882.5	$Q(5)$	56 368 376.6(100)	4.7
		-3115.0	$Q(6)$	56 368 144.1(100)	1.7
		-3381.2	$Q(7)$	56 367 877.9(100)	3.2
		-3689.4	$Q(8)$	56 367 569.7(100)	0.8
		-4032.7	$Q(9)$	56 367 226.4(100)	1.5
		-4414.1	$Q(10)$	56 366 845.0(100)	2.2
		-4826.2	$Q(11)$	56 366 432.9(150)	10.3
		-6322.6	$Q(14)$	56 364 936.5(100)	2.7
	56 371 258.2	-6898.2	$Q(15)$	56 364 360.0(50)	-1.6
		-7508.1	$Q(16)$	56 363 750.1(50)	-1.4
		-8152.0	$Q(17)$	56 363 106.2(50)	2.6
$P_6(19)$	57 276 297.7	-1010.4	$R(38)$	57 275 287.3(70)	+0.1
$P_6(18)$	57 403 401.8	-1599.6	$R(44)$	57 401 802.2(80)	-2.4

<sup>a</sup> The CO transitions are designated by  $\nu''$  and  $J''$ , the lower-state quantum numbers.

<sup>b</sup> The estimated uncertainty in the last digits is given in parentheses.

**Table 2. Heterodyne-Frequency Measurements on Nitrous Oxide at 9  $\mu\text{m}$** 

Reference Laser, $^{12}\text{C}^{18}\text{O}_2$ Transition	$\text{N}_2\text{O}$ Transition, Rot.-Vib.	$\text{N}_2\text{O} - \text{CO}_2$ Difference Frequency (MHz)	Measured $\text{N}_2\text{O}$ Frequency (MHz) <sup>a</sup>
$R_{\text{II}}(44)$	$P(65) 03^1f_0-01^1f_0$	-1508.3	33 271 867.2(100)
$R_{\text{II}}(42)$	$P(48) 05^1e_0-03^1e_0$	1344.2	33 246 066.3(100)
	$P(55) 04^2f_0-02^2f_0$	-385.0	33 244 337.1(100)
	$P(75) 02^0_0-00^0_0^b$	-1302.3	33 243 419.8(100)
	$P(63) 03^1e_0-01^1e_0$	-3262.4	33 241 459.7(60)
$R_{\text{II}}(40)$	$P(64) 03^1e_0-01^1e_0$	1805.6	33 217 307.7(60)
	$P(51) 05^1f_0-03^1f_0$	449.3	33 215 951.4(60)
$R_{\text{II}}(38)$	$P(58) 04^0_0-02^0_0$	2252.7	33 187 966.2(60)
	$P(69) 03^1f_0-01^1f_0$	-1326.3	33 184 387.2(60)
	$P(59) 04^2e_0-02^2e_0$	-3343.2	33 182 370.3(150)
$R_{\text{II}}(34)$	$P(55) 05^1f_0-03^1f_0$	-858.1	33 123 564.8(60)
	$P(53) 05^1e_0-03^1e_0$	-1872.0	33 122 550.9(100)

<sup>a</sup> The uncertainty in the last digits is given in parentheses.

<sup>b</sup> We believe that the weaker transition  $05^3f_0-03^3f_0$  also occurs at this frequency.

the 5.3- $\mu\text{m}$  measurements. For the 9.0- $\mu\text{m}$  measurements, the pressures ranged from 1.33 to 2.66 kPa (10 to 20 Torr), and the cell was heated to about 150°C.

## RESULTS AND DESCRIPTION OF THE ANALYSIS

The frequency measurements and line assignments in the 9- $\mu\text{m}$  region are given in Table 2. The assignments were determined by combining calculated transition frequencies using constants taken from Refs. 8, 9, 15, 16 with the observed Fourier-transform spectrum of a 10-m path of 1.33 kPa (10 Torr) of  $\text{N}_2\text{O}$  at 380 K. Since only a few high- $J$  transitions were observed for each vibrational transition, we have not attempted to incorporate the present measurements in a reanalysis of the  $\text{N}_2\text{O}$  data for the 9- $\mu\text{m}$  region.

The frequency measurements and the line assignments in the 5.3- $\mu\text{m}$  region are given in Table 1. The measurements given by Amiot and Guelachvili and reported in Table 7 of Ref. 6 were used to assign the present measurements. Both the analysis of the present data and the calculation of the transition frequencies used the following energy equations:

$$F(00^0_0) = B_0J(J+1) - D_0J^2(J+1)^2 + H_0J^3(J+1)^3, \quad (1)$$

$$F(01^1_1) = \nu_0 + B_{011}[J(J+1)] - D_{011}[J(J+1) - 1]^2 + H_{011}[J(J+1) - 1]^3 \pm 0.5\{q - q_J[J(J+1)]\}J(J+1), \quad (2)$$

and

$$\nu_{\text{obs}} = F(01^1_1) - F(00^0_0). \quad (3)$$

The ground-state constants used in this analysis were taken from Ref. 5 since these constants resulted from a fit that combined microwave measurements with the Fourier-transform spectroscopy (FTS) measurements of Refs. 6, 9, and 16-18.

The present measurements were used to determine the upper-state constants given in Eq. (2). The microwave measurements by Andreev *et al.*<sup>19</sup> were included in the analysis to define the rotational constants better. Since the present  $Q$ -branch measurements do not extend to high- $J$

values, the  $Q$ -branch measurements of Amiot and Guelachvili<sup>6</sup> were included in the fit, although their FTS measurements were allowed to define a different band center. Since the FTS  $P$ - and  $R$ -branch measurements covered a wider frequency range and therefore were more likely to be subject to systematic frequency-calibration problems, we did not include the FTS  $P$ - and  $R$ -branch data in this analysis. Table 3 gives the constants resulting from the present analysis. Table 4 gives the calculated  $\text{N}_2\text{O}$  line frequencies given by the constants of Table 3 and the uncertainties that resulted from the variance-covariance matrix obtained for the upper-state constants (plus a pressure-shift uncertainty of 2 MHz).

Our analysis of the  $Q$ -branch data of Amiot and Guelachvili<sup>6</sup> agrees with their reported band center. The band center given by the present heterodyne measurements is about 4 MHz higher than the band center given by Amiot and Guelachvili. The direction of the disagreement was somewhat surprising to us since previous comparisons had indicated that the FTS measurements were always too high.<sup>4,5</sup>

The present measurements were made at pressures ranging from 0.5 kPa (4 Torr) (for the  $Q$  branch) to 2 kPa (15 Torr) (for the  $P$  and  $R$  branches). Since the pressure-induced shift measured for two bands near 3700  $\text{cm}^{-1}$  was about -1.12 kHz/Pa (-0.15 MHz/Torr),<sup>5</sup> we believe that the shift coefficient will be smaller than -1.5 kHz/Pa (-0.2 MHz/Torr) for the newly measured band. This coefficient would result in the present measurements' being too low by less than 0.8 MHz

**Table 3. Rovibrational Constants for the  $01^1_1-00^0_0$  Band of  $\text{N}_2\text{O}$** 

$\nu_0$ (MHz)	56 368 945.7(30) <sup>a</sup>
$B(01^1_1)$ (MHz)	12 528.8778(25)
$D(01^1_1)$ (kHz)	5.1954(38)
$H(01^1_1)$ (mHz)	4.92(112)
$q(01^1_1)$ (MHz)	27.22611(214)
$q_J(01^1_1)$ (Hz)	-95.91(228)
$B_0$ (MHz)	12 561.633 60 <sup>b</sup>
$D_0$ (kHz)	5.278 421
$H_0$ (mHz)	-0.5152

<sup>a</sup> The uncertainty (twice the estimated standard error) in the last digits is given in parentheses.

<sup>b</sup> The ground-state constants were taken from Ref. 5.

**Table 4. Wave Numbers ( $\text{cm}^{-1}$ ) Calculated<sup>a</sup> for the  $01^11-00^00$  Band of  $\text{N}_2\text{O}$** 

$J''$	P Branch	R Branch	Q Branch	$J''$
0	—	1881.100 57(12)	—	0
1	—	1881.932 40(12)	1880.264 36(12)	1
2	1878.586 51(12)	1882.761 13(12)	1880.261 81(12)	2
3	1877.742 31(12)	1883.586 75(12)	1880.257 98(12)	3
4	1876.895 04(12)	1884.409 27(12)	1880.252 88(12)	4
5	1876.044 69(12)	1885.228 67(12)	1880.246 50(12)	5
6	1875.191 27(12)	1886.044 96(12)	1880.238 84(12)	6
7	1874.334 78(12)	1886.858 12(12)	1880.229 91(12)	7
8	1873.475 23(12)	1887.668 16(12)	1880.219 71(12)	8
9	1872.612 62(12)	1888.475 07(12)	1880.208 24(12)	9
10	1871.746 96(12)	1889.278 84(12)	1880.195 49(12)	10
11	1870.878 25(12)	1890.079 48(12)	1880.181 47(12)	11
12	1870.006 49(12)	1890.876 98(12)	1880.166 19(12)	12
13	1869.131 70(12)	1891.671 33(12)	1880.149 63(12)	13
14	1868.253 87(12)	1892.462 54(12)	1880.131 81(12)	14
15	1867.373 00(12)	1893.250 59(12)	1880.112 73(12)	15
16	1866.489 12(12)	1894.035 48(12)	1880.092 38(12)	16
17	1865.602 21(12)	1894.817 22(12)	1880.070 77(12)	17
18	1864.712 28(12)	1895.595 79(12)	1880.047 90(12)	18
19	1863.819 34(12)	1896.371 20(12)	1880.023 77(12)	19
20	1862.923 40(12)	1897.143 43(12)	1879.998 38(12)	20
21	1862.024 45(12)	1897.912 49(12)	1879.971 75(12)	21
22	1861.122 51(12)	1898.678 37(12)	1879.943 86(12)	22
23	1860.217 57(12)	1899.441 08(12)	1879.914 72(12)	23
24	1859.309 65(12)	1900.200 60(12)	1879.884 34(12)	24
25	1858.398 74(12)	1900.956 93(12)	1879.852 71(12)	25
26	1857.484 86(12)	1901.710 07(12)	1879.819 85(12)	26
27	1856.568 01(12)	1902.460 02(12)	1879.785 74(12)	27
28	1855.648 19(12)	1903.206 78(12)	1879.750 41(12)	28
29	1854.725 41(12)	1903.950 33(12)	1879.713 84(12)	29
30	1853.799 67(12)	1904.690 68(12)	1879.676 05(12)	30
31	1852.870 99(12)	1905.427 83(12)	1879.637 03(12)	31
32	1851.939 36(12)	1906.161 77(12)	1879.596 80(12)	32
33	1851.004 80(12)	1906.892 50(12)	1879.555 35(12)	33
34	1850.067 30(12)	1907.620 02(12)	1879.512 69(12)	34
35	1849.126 87(12)	1908.344 33(12)	1879.468 82(13)	35
36	1848.183 53(12)	1909.065 42(12)	1879.423 75(13)	36
37	1847.237 27(12)	1909.783 29(12)	1879.377 49(13)	37
38	1846.288 10(12)	1910.497 93(12)	1879.330 03(13)	38
39	1845.336 03(12)	1911.209 36(12)	1879.281 39(13)	39
40	1844.381 07(12)	1911.917 56(13)	1879.231 56(13)	40
41	1843.423 22(12)	1912.622 54(13)	1879.180 56(14)	41
42	1842.462 48(13)	1913.324 29(14)	1879.128 39(14)	42
43	1841.498 87(13)	1914.022 81(14)	1879.075 05(14)	43
44	1840.532 40(14)	1914.718 10(15)	1879.020 56(14)	44
45	1839.563 06(14)	1915.410 16(16)	1878.964 92(14)	45
46	1838.590 87(15)	1916.098 99(17)	1878.908 13(14)	46
47	1837.615 83(16)	1916.784 59(18)	1878.850 20(15)	47
48	1836.637 96(17)	1917.466 95(19)	1878.791 15(15)	48
49	1835.657 25(18)	1918.146 08(21)	1878.730 97(15)	49
50	1834.673 73(19)	1918.821 98(22)	1878.669 67(15)	50

<sup>a</sup> The uncertainty in the last digits (one standard error) is given in parentheses.

for the Q branch or up to 3 MHz for the P and R branches. This may account for the observed small systematic deviations in the fit of these measurements. The uncertainty contributed by the pressure-shift uncertainty is included in the uncertainties given in Table 4 and would tend to make the values given in that table too low by a small amount. We did

not attempt to correct any of the data or calculations for the possible effect of pressure shifts.

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