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Final Report

TRANSMISSIVITY OF CARBON MONOXIDE

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## Abstract

The line strengths and self & N<sub>2</sub> broadened half-widths for selected lines of the 4.6 $\mu$ m fundamental band of carbon monoxide have been determined. The band strength determined  $281 \pm 14 \text{ cm}^{-1} (\text{atm. cm.})^{-1}$  at stp. is higher than previously reported measurements. The half widths agree well with other measurements and calculations in the literature.

## 1. Introduction

A detailed knowledge of the carbon monoxide spectrum is important in such fields as Astrophysics, Chemistry, Combustion, Meteorology, Molecular Spectroscopy, Planetary Atmosphere Studies, Radiation thermometry, etc.

There has been a considerable amount of research to develop a method to determine accurately the global distribution of CO from a satellite measurement. (Grenda et al. 1971) In such a measurement the radiation in the region of the electromagnetic spectrum where CO has absorption bands, (and preferably where no other constituents have absorption bands) emanating from the earth is measured. A prior detailed knowledge of these absorption bands is required for the calculation of the atmospheric transmissivity. Molecular absorption bands contain many hundreds of individual absorption lines, some strong and some weak. If the line parameters (line strength and line half-width) are known for each line in the absorption bands the atmospheric transmissivity can be accurately calculated. Simplified calculations of transmissivity would result if band models could be used.

The infrared spectrum of CO has been extensively studied because of its great simplicity. Detailed theoretical calculations of the CO spectra have been made by many authors. (Chackerian 1970, Kunde 1968, Young 1968) Many experimental determinations of line strengths and widths and line shapes have been reported in the literature. Unfortunately due to the inherent difficulty of making such measurements there is a considerable variation among the results obtained by investigators. This report presents the results of experimental measurements conducted at high resolution ( $0.05\text{-}0.08\text{ cm}^{-1}$ ) of the carbon monoxide  $4.6\mu\text{m}$  absorption spectrum. The experimental results are compared with theoretical calculations.

## 2. Experimental

The experimental measurements were conducted using a Jarrell Ash 1.83 meter Fastie Ebert vacuum scanning spectrometer. The detailed description of the apparatus and the measurement technique have been described by Chaney (1972). A graphite rod mounted inside a water cooled housing and heated electrically was used as the source. The sample cell was 8.74 cm. and 4.0 cm. in inside diameter. An indium antimonide detector mounted in an IR-15 dewar was used. The output signal from the detector after passing through pre-amplifier and a phase lock amplifier is a D.C. signal varying from 0 to  $\pm 5$  volts and is recorded on a Leeds and Northrup Analog chart recorder. The analog signal was digitized by a Hewlett-Packard DY 2401 C Integrating Voltmeter which was interfaced through an H. P. 2526 cardcoupler to an IBM 526 Summary Punch and the data was punched on IBM cards. Each data point corresponded to 0.1 counter number or drum turn (approximately  $0.008 \text{ cm}^{-1}$ ). The data scan rate was approximately 220 seconds per wavenumber. For most scans a signal to noise ratio of approximately 200:1 was obtained, and a time constant of 0.1 seconds was used.

The gas samples used were from commercial cylinders made by Matheson Co. and had a stated purity of 99.5% or higher. The isotopic composition of the CO samples was assumed to be close to that of atmospheric CO of  $\text{C}^{12}\text{O}^{16}$  (98.654%),  $\text{C}^{13}\text{O}^{16}$  (1.106%),  $\text{C}^{12}\text{O}^{18}$  (.202%),  $\text{C}^{12}\text{O}^{17}$  (.037%) and  $\text{C}^{13}\text{O}^{18}$  (.002%).

The pressures in the sample cell were measured with a Baratron pressure gauge to 0.01 mm. of Hg. accuracy. Room temperature during a scan was maintained to  $\pm 0.5^\circ \text{C}$ . The cell temperatures ranged from  $295^\circ$  to  $301^\circ \text{K}$  for all our measurements and during a scan the cell temperature did not vary significantly.

The 4.6  $\mu\text{m}$  band was studied using 2 separate gratings. The first grating was blazed at 16 $\mu\text{m}$  and used in the 4th order. The second grating was blazed at 4 $\mu\text{m}$  and used in the order. The measured resolutions for the two gratings were approximately 0.05 and 0.08  $\text{cm}^{-1}$  respectively. Six regions of the 4.6 $\mu\text{m}$  band of CO were scanned. (Table 1)

TABLE 1

Regions of the band studied

(unless otherwise specified the 1st grating was used)

Region	Wavenumber( $\text{cm}^{-1}$ )	$\text{C}^{12}\text{O}^{16}$ Lines
1	2055-2065	P19-P20
2	2090-2095*	P12
3	2135-2150	R1-P2
4	2180-2185	R10
5	2213-2217	R20
6	2224-2230	R23-R24

\* 2nd grating

These regions were selected for study because of the relative freedom from overlapping of lines.

The spaces between the chopper and the cell ( $\sim 2$  cm. long) and between the cell and entrance window ( $\sim 2$  cm. long) were enclosed with plastic and tape and continuously purged with the boil off from a liquid nitrogen storage tank. However for low pressure data, where the equivalent width of a line was small it was noticed that a small extraneous absorption (in the worst possible case approximately 10% of the equivalent width at 10 mm of Hg. of CO.) led to erroneous results for line strengths and widths. This was found to be due to the presence of additional CO (produced by cooling water leaking onto the hot graphite source) in the optical path. This additional absorption could also be noticed on some background spectra. Later measurements were greatly improved by having a greater flow rate of nitrogen for the purge.

Some original nitrogen broadened data was also found unuseable because the technique, (gases mixed in the manifold and valved into the sample cell) used in filling the sample cell was not satisfactory. This error was noticed in the spectra, where even for strong lines for the same path length of carbon monoxide, and increasing amounts of the broadening gas nitrogen, the equivalent width randomly fluctuated and sometimes decreased considerably. The method of inletting of the gases was improved and sufficient time for mixing of the gases was allowed (Chaney 1972).

### 3. Analysis

The wavenumbers of the carbon monoxide lines are accurately known (Benedict et al 1962). Using these as calibration it was found that the relation between drum turn and wavelength was almost linear over wavenumber ranges as large as  $100 \text{ cm}^{-1}$ . Thus the drum turns could be converted into wavenumbers using a linear interpolation with wavelength between the CO lines. The spectra were also normalized. As many authors discuss (Korb et al 1968) the determination of the 100% and 0% transmission lines is highly susceptible to error. A 100% transmission scan was originally taken, but it was found that this line fluctuated considerably in amplitude due possibly to the variation of transmission through the order sorting interference filter because of temperature variations. The 100% transmission line was therefore taken as a straight line connecting the spectral regions on each side of absorption line where the absorption appeared small. A wing and base correction (described later) must also be applied. By blocking the beam a 0% transmission line was noted and this was found to be a constant value dependent only on the amplifier gain setting. This 0% line was also obtained from the saturation region of strong lines. The spectra were normalized by using the relation  $(T_{100} - T_x) / (T_{100} - T_0)$ .

The method of curve of growth was used in the determination line strengths and widths. It was found convenient to separate the data into individual spectral lines for the calculation of the equivalent width. Penner (1959) has shown that the equivalent width is invariant with respect to the spectral slitwidth. For each spectral line the values of equivalent width for the different CO and N<sub>2</sub> pressures were calculated using the trapezoidal rule. Where there was weak blending by a neighboring isotopic line a correction for the blending was made by subtracting an area equal to the product of the blending line strength and the absorber path length from the total equivalent width.

An iterative procedure was then carried out for the calculation of the line parameters. An initial guess of line strength S and self- and nitrogen-broadened half-widths  $\alpha_{\text{CO}}$  and  $\alpha_{\text{N}_2}$  were made. The values of the parameters a,  $x_D$  and  $x_L$  were calculated.

$$a = (\gamma_L / \gamma_D) \cdot (\ln 2)^{1/2}, \quad x_D = (\ln 2 / \pi)^{1/2} \cdot (Su / \gamma_D), \quad x_L = Su / 2\pi\gamma_L$$
where  $\gamma_D$  is the Doppler half-width ( $= 3.58 \cdot 10^{-7} (T/M)^{1/2} \nu$ ) cm<sup>-1</sup> and  $\gamma_L$  is the Lorentz half-width ( $= \alpha_{\text{CO}} p_{\text{CO}} + \alpha_{\text{N}_2} p_{\text{N}_2}$ ) cm<sup>-1</sup>). The values of the equivalent width were corrected to include the wing and base effects. The correction has been given by Korb et. al. (1968).

$$W_v = W_{\text{meas}} + \frac{4 Su}{\pi} \frac{\gamma_L}{\eta} \left[ 1 + \frac{2}{3} d_1 \left( \frac{\gamma_L}{\eta} \right)^2 + \frac{3}{5} d_2 \left( \frac{\gamma_L}{\eta} \right)^4 \right]$$

$$\text{where } d_1 = \frac{3}{2} a^{-2} - 1, \quad d_2 = \frac{15}{4} a^{-2} - 5a^{-2} + 1.$$

and  $\eta$  is the distance from the line center to where the experimental measurement of the area was carried out, and u is the absorber path length.

$$u = p_{\text{CO}} \cdot L \cdot (273/T) \text{ (atm - cm) STP}$$

The wing and base corrected equivalent widths were corrected for Doppler broadening of the spectral line. Corrections for a Voigt line given by

Yamada (1968) in a detailed paper were used.

1.  $a \geq 0.5$   $W_L = W_V$
2.  $0 < a < 0.5$   $W_L = W_V / \left[ 1 + \frac{3}{16} a^{-2} x_L^{-1} + \frac{45}{512} a^{-4} x_L^{-2} + \dots \right]$  for  $W_L > W_D$   
 and  $W_D = W_V / \left[ 1 + \sqrt{a x_D} / 2\pi^{n/2} \right]$  for  $W_D > W_L$

where  $n$  is specified by the order of magnitude of  $a$  ( $= 10^{-n+1}$ ). The subscripts L, D and V refer to Lorentz, Doppler and Voigt respectively. We had a few measurements at extremely low pressures where  $W_D > W_L$  and the line strength could be directly determined from  $W_D$ . The line strengths calculated were however inaccurate because of CO from the source present in the optical path.

For most of the data it was found that  $W_L$  was much greater than  $W_D$ . The corrected equivalent widths  $W_L$  were calculated. The quantity  $W_L / p_T$  was calculated and its approximate constancy for a given  $x_L$  showed that the assumption of Lorentz line shape is accurate for our measurements. Benedict et. al. (1962) have discussed a modified Lorentz shape, where beyond  $4 \text{ cm}^{-1}$  from the line center, the line shape is given by multiplying the Lorentz expression by a shape factor given by

$$X = \exp \left[ -0.015 (|\nu - \nu_0| - 4) \right].$$

Burch and Gryvnak (1967) have shown that the extreme wings of the lines are sub-lorentzian. Our measurements show general agreement with their results, in that deviations near the line centers are small.

The Ladenberg and Reiche function  $f$  was calculated for each data scan  $f = x_L \cdot e^{-x_L} \left[ J_0(ix_L) - iJ_1(ix_L) \right]$

The data for which  $f \leq 0.5$  was used to calculate the line strength from

$$S = \frac{W_L \cdot x_L}{11 \cdot f} \text{ cm}^{-1} / (\text{atm} \cdot \text{cm}) \text{ STP}$$

The reason for taking  $f < 0.5$  is that for this case  $x_L/f$  is close to unity and so the value of  $S$  calculated is not too highly dependent on the value of  $x_L$ , i.e. on the initial guesses of strength and half-width. The value of 0.5 is arbitrary. The self-broadened data for which  $f > 0.5$  was used to calculate the self-broadened half-width  $\alpha_{CO} = W_L / (2\pi p_T \cdot f) \text{ cm}^{-1} \cdot \text{atm}^{-1}$

The nitrogen broadened data for which  $f > 0.5$  was used to calculate the nitrogen-broadened half-width.

$$\gamma_L = W_L / (2\pi \cdot f) \text{ cm}^{-1} \quad \text{and} \quad \alpha_{N_2} = \frac{(\gamma_L - \alpha_{CO} p_{CO})}{p_{N_2}} \text{ cm}^{-1} \text{ atm}^{-1}$$

Averages of the new calculated parameters  $S$ ,  $\alpha_{CO}$  and  $\alpha_{N_2}$  were taken. Having obtained these new values for the line parameters  $S$ ,  $\alpha_{CO}$  and  $\alpha_{N_2}$  the calculations were repeated using these new values for the initial guess. The calculations were repeated till a convergence criteria was met. We carried out the iterations till the difference between the calculated values and the guessed values (previously calculated) differed only by .0002, i.e. well within the estimated experimental error.

Due to our limited data, 4 spectral lines P12, R1, R10 and R20 were analyzed first. The line strengths of these 4 spectral lines were used to calculate the band strength of the fundamental of  $C^{12}O^{16}$  using the Herman Wallis (1955) formula.

$$S_m = \frac{S_{\text{band}} |m| (\nu / \nu_c) F_m}{Q} \exp \left[ \frac{-B \cdot m (m-1) hc}{kT} \right]$$

where the Herman Wallis factor  $F_m = 1$  for the fundamental (Young and Eachus 1966). From our band strength we generated the line strengths of the other  $C^{12}O^{16}$  lines we have studied. Using these line strengths, and adopting a similar iterative procedure (which converged rapidly) as described above the line widths  $\alpha_{CO}$  and  $\alpha_{N_2}$  were calculated from the equivalent width measurements in self and nitrogen broadening respectively, for all  $f$  values.

$$\gamma_L = W_L / (2\pi f) \text{ cm}^{-1}$$



We have determined the line intensities of the other isotopic lines and calculated the Band intensities of the fundamentals of the isotopic species  $C^{13}O^{16}$ ,  $C^{12}O^{18}$  and  $C^{12}O^{17}$ .

#### 4. Results and Discussion

The line strengths and line widths of the P12, R1, R10 and R20 were calculated and are tabulated in Table 4. The Band strength for the fundamental of  $C^{12}O^{16}$  calculated from these individual measurements is shown in Table 2.

TABLE 2

Band strength from individual measurements

Line	Line strength $cm^{-1}(atm. cm)^{-1}stp$	Band strength $cm^{-1}(atm cm)^{-1}stp$	Standard deviation %
P12	7.09	279.9	2.8
R1	5.35	296.6	2.7
R10	10.36	279.2	2.2
R20	1.11	275.4	1.0

We did not use the band strength obtained from the R1 line measurement in calculating the average band strength because of the blending of the line with the isotopic  $C^{13}O^{16}$  R15 line. The average Band strength from the other three measurements gives a value  $278.2 cm^{-1}(atm. cm.)^{-1}stp$ . Line strengths for the other  $C^{12}O^{16}$  lines studied were calculated from this value of band strength and shown in Table 4. The line half-widths were calculated and are tabulated in Table 4. From our measurements the calculations of the average band strengths for the fundamentals of  $C^{13}O^{16}$ ,  $C^{12}O^{18}$  and  $C^{12}O^{17}$  are 2.7, 0.44, 0.1  $cm^{-1}(atm. cm.)^{-1}stp$ . The total band strength of the fundamentals is estimated as  $281 \pm 14 cm^{-1}(atm-cm)^{-1}STP$ . The hot bands are less than  $0.1 atm^{-1} cm^{-2}$  in strength at this temperature.

Table 3 lists other measurements of the  $4.6 \mu m$ . band intensity of CO.

TABLE 3

Band Intensities of the 4.6  $\mu\text{m}$  band of CO

Year	Author	Band Intensity $\text{cm}^{-1}(\text{atm. cm})^{-1} \text{stp.}$
1951	Penner and Weber	$260 \pm 12$
1953	Locke and Herzberg	245
1953	Vincent-Geisse	262
1956	Shaw and France	238
1962	Benedict et al	236
1962	Burch and Williams	$260 \pm 20$
1965	Armstrong and Welsh	$258 \pm 4$
1966	Hochard de Molliere	247.8
1973	Present Study	$281 \pm 14$

Figure 1 shows our measurements of the self-broadened line widths compared with other recent measurements. The results of this work agree very well with the direct measurements of Kostkowski and Bass (1961). The nitrogen broadened half-widths are plotted in Figure 2 and compared with the theoretical calculations of Bouanich and Haeusler (1972). Our results show fair agreement with the results of other authors.

Our total band intensity is approximately 8% higher than the mean value of  $260 \text{ cm}^{-1}(\text{atm. cm})^{-1} \text{STP}$  of some of the more recent determinations. We estimate that without the inclusion of the base correction in our analysis we would have obtained a band intensity about 8% lower. This may be responsible for some of the difference between our results and those of the authors in Table 3.

The errors in this type of measurement have been very well summarized by Reichle (1969). The mathematical errors of truncation and round off were negligible. The wing and base area correction contributed 18% of the total area in the most extreme case, but was usually half or less of this value. The error in estimating this correction was much less than the error

TABLE 4

Measured Line Intensities and Half-Widths

<u>Line</u>	<u>Isotope</u>	<u>Line Center</u>	$S \text{ cm}^{-1}$ $[\text{atm}^{-1} \text{ cm}^{-1}]$ <u>STP</u>	$\alpha$ CO $\text{atm}^{-1} \text{ cm}^{-1}$	$\alpha$ N <sub>2</sub> $\text{atm}^{-1} \text{ cm}^{-1}$
P20	26	2059.9	1.0	.060	-
P19	26	2064.4	1.39	.063	-
P12	26	2094.9	7.09	.071	.064
R11	28	2133.3	0.016	-	-
P 2	26	2135.5	4.89	.089	-
R11	36	2137.6	0.0875	-	-
P 1	26	2139.4	2.54	.089	-
R12	36	2140.8	0.085	-	-
R14	28	2143.0	0.010	-	-
R13	36	2144.0	0.072	-	-
R 7	27	2145.1	0.0045	-	-
R 0	26	2147.1	2.6	.087	-
R16	28	2049.3	0.006	-	-
R 1	26	2150.9	5.35	.089	.081
R10	26	2183.2	10.36	.070	.064
R20	26	2215.7	1.11	.055	.048
R23	26	2224.7	.38	.048	-
R24	26	2227.6	.26	.051	-

\*The notation used to identify the isotope is: 26 = C<sup>12</sup>O<sup>16</sup>, 36 = C<sup>13</sup>O<sup>16</sup>,  
28 = C<sup>12</sup>O<sup>18</sup>, and 27 = C<sup>12</sup>O<sup>17</sup>

involved in estimating the background. In addition to the small random fluctuations which were minimized by taking the average of 30 data points on either side of the spectral line, there were systematic shifts in the background level from scan to scan and presumably during each scan. The shifts between scans were systematic and thought to be associated with a slow warming in some part of the instrument. They amounted to about a 5% change during a 4 hour period and hence would be much less than 1% during the time taken to scan an individual line. In addition to the background fluctuations discussed above, some of the spectra showed depressions or enhancements of the background level over frequency intervals of a few tenths of a wave-number of an amplitude of about 1% although usually the amplitude was much less (for e. g. see figure 5). These appear to be the largest source of error in estimating the backgrounds. It is difficult to assess the effect on the final results, but we estimate an error of 2% or less in the equivalent widths for most of the data.

The error due to source contamination contributed at worst an error of 8% of the equivalent width for measurements of 10mm Hg of CO self-broadened data, for a few of the first measurements. This error was much smaller for later data where a greater flow rate of nitrogen purge was used. Errors in the measurement of temperature, pressure, composition, path length are less than 0.5% and do not contribute to any significant error in the calculations. The error in the digitizing system was less than 0.1%. Shift of wavelength marking, due to variation in temperature, was less than 0.5% (see fig. 5). We estimate that the error in the measurement of the band strength is  $\pm 5\%$ . The error in the measurement of line widths are shown as error bars on Figures 1 & 2. The error bars are the standard deviation of the individual half-widths measurements. Figures 3, 4, 5 show sample spectra obtained. The typical signal to noise ratio obtained is depicted.

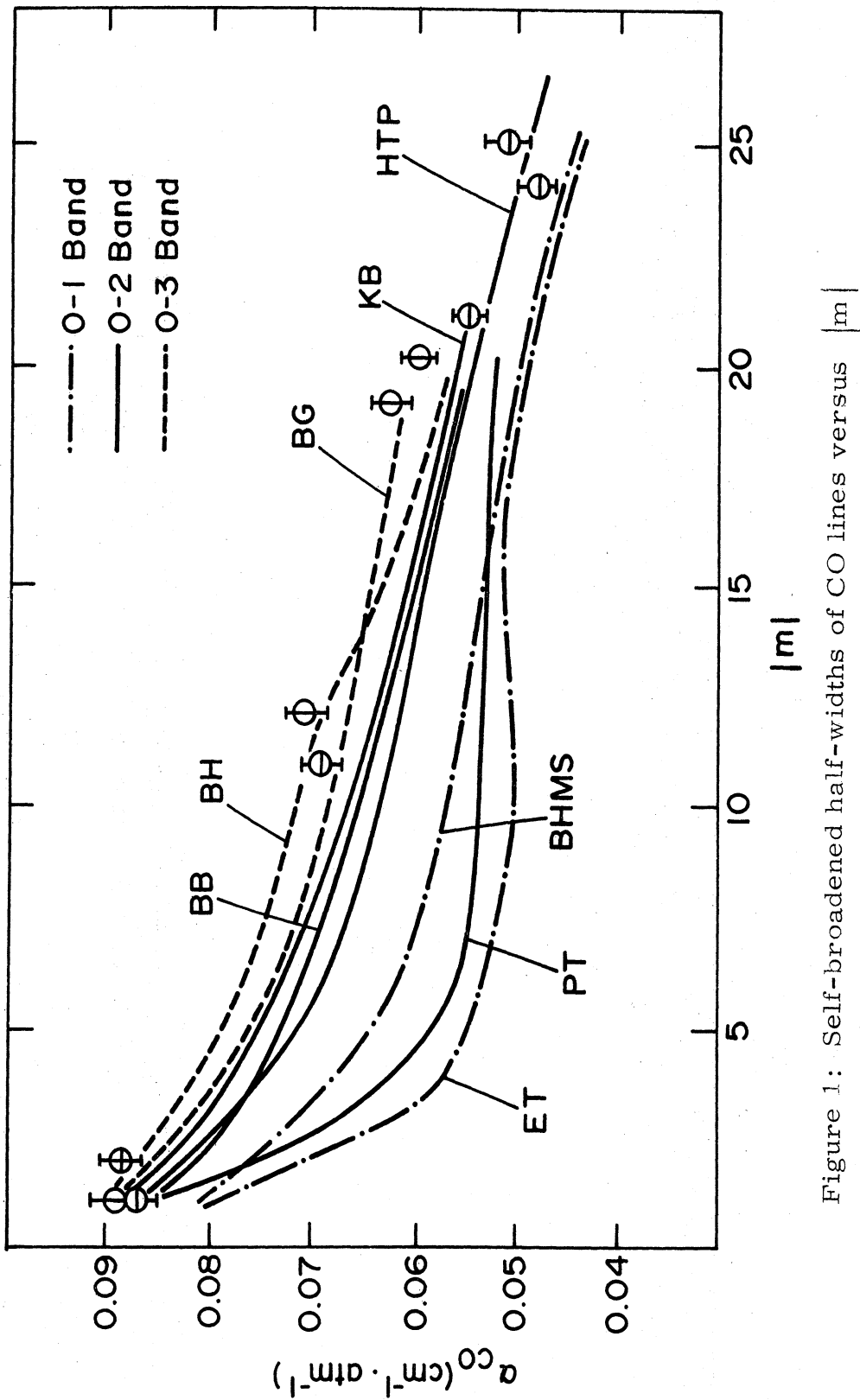


Figure 1: Self-broadened half-widths of CO lines versus  $|m|$ . Results of this work ( $\Phi$ -P branch and  $\Theta$ -R branch) are shown along with results in the 0-1 band of Eaton and Thompson (1959)-ET; Benedict et. al. (1962)-BHMS; 0-2 band of Kostkowski and Bass (1961)-KB; Plyler and Thiobault (1963)-PT; Hunt et. al. (1968)-HTP; Bouanich and Brodbeck (1973)-BB; 0-3 band of Burch and Gryvnak (1967)-BG; Bouanich and Haeusler (1972)-BH.

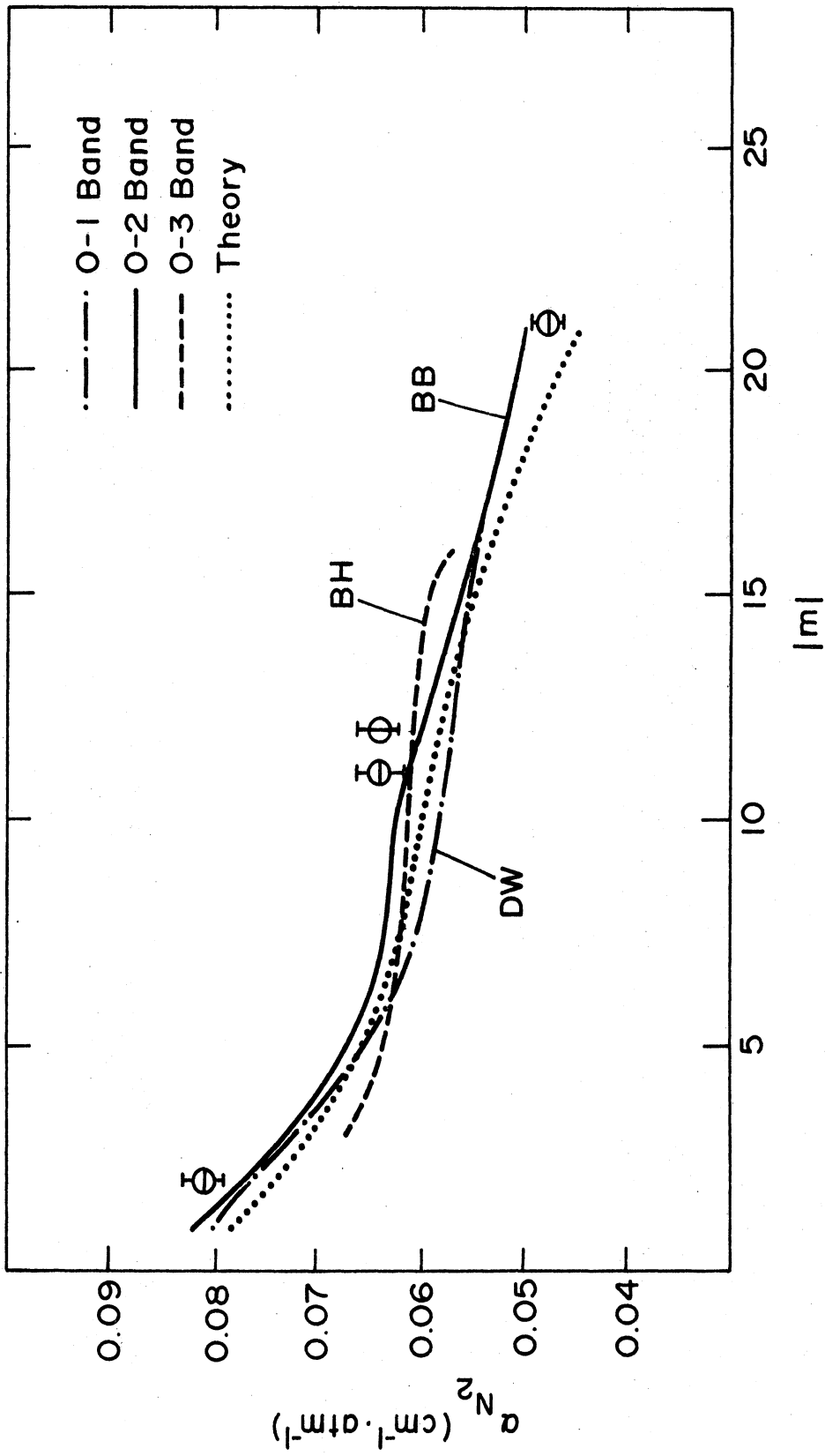


Figure 2: Nitrogen Broadened half-widths of CO lines versus  $|m|$ . Results of this work ( $\Phi$ , P branch and  $\Theta$ , R branch) are shown with the theoretical ( $\dots$ ) and experimental (0-3 band) results of Bouanich and Haeusler (1972). - BH; and results of Draegert and Williams (1968) (0-1 band) -DW; and Bouanich and Brodbeck (1973) (0-2 band) -BB.

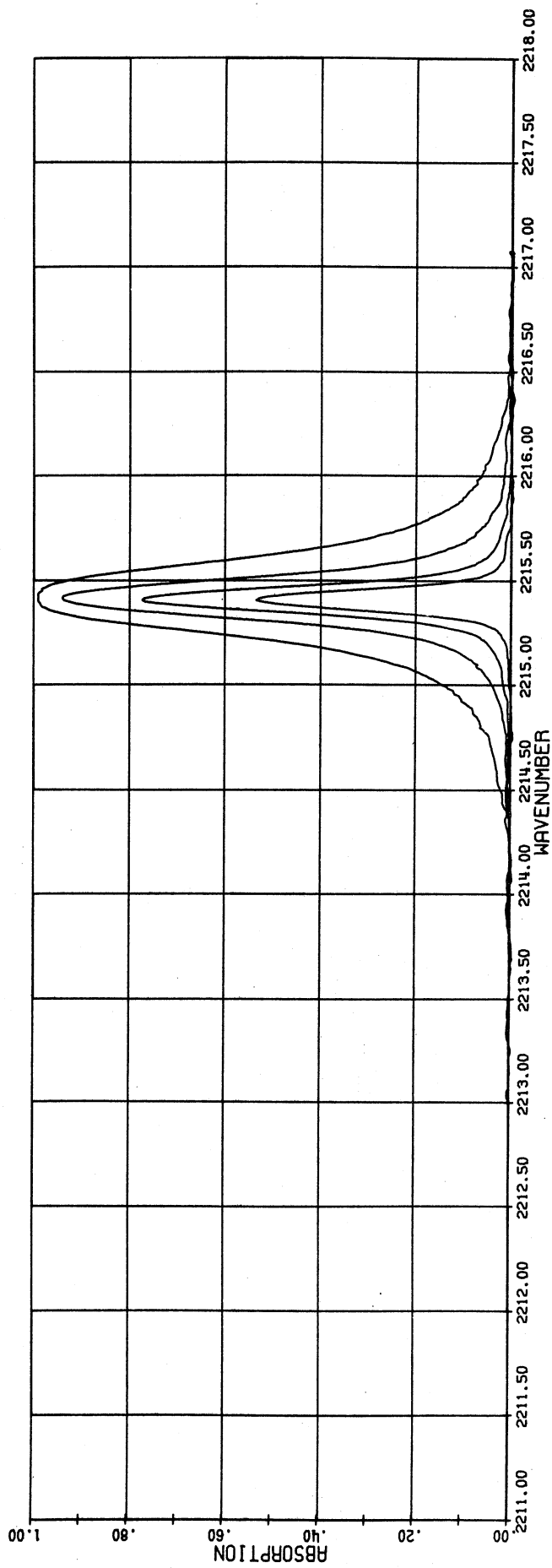


Fig. 3 Spectra of the R20 line of  $C^{12}O^{16}$  self-broadening study

Scans . #519, 520, 521 and 522.

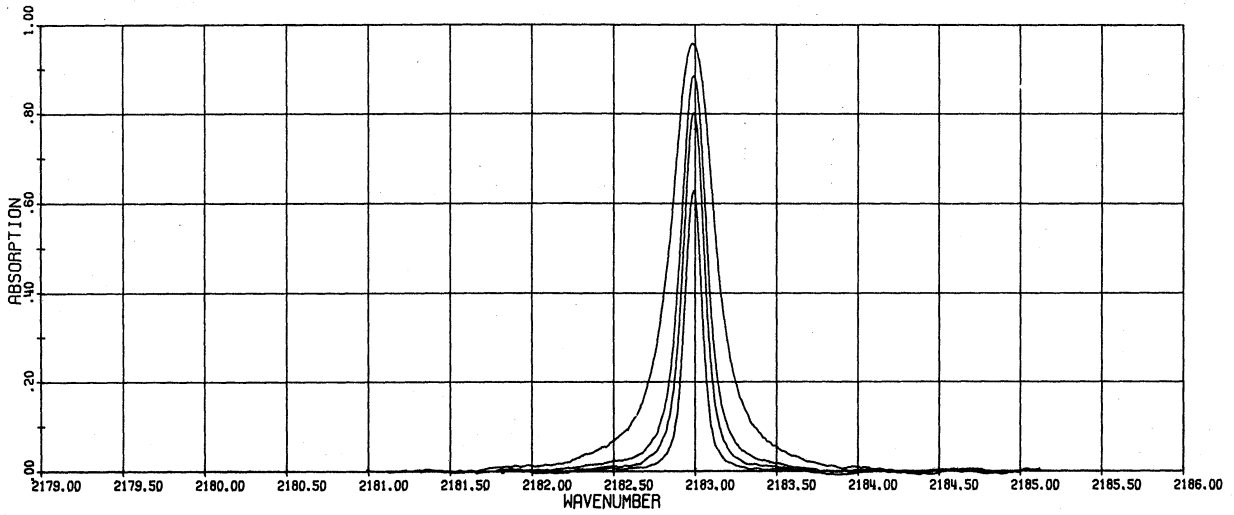


Fig. 4. Spectra of the R10 line of  $C^{12}O^{16}$  nitrogen broadening study  
Scans. # 502, 504, 505 and 507.

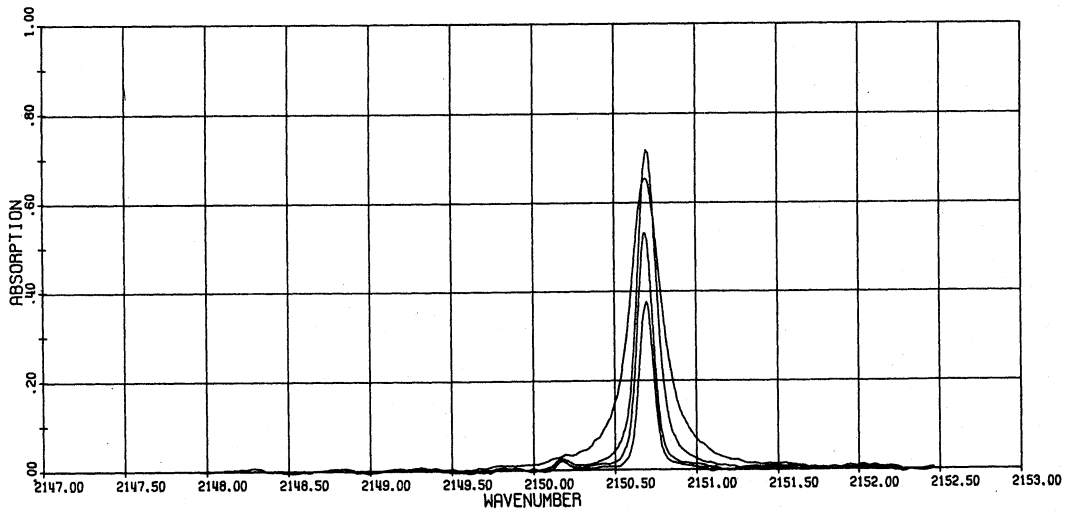


Fig. 5. Spectra of the R1 line of  $C^{12}O^{16}$  nitrogen broadening study.  
Scans. # 510, 512, 514 and 516.



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APPENDIX: Data Summary

Scan No.	Wave Number cm <sup>-1</sup>	Lines C <sup>12</sup> O <sup>16</sup>	Pressure mm. Hg.		Remarks
			CO	N <sub>2</sub>	
311-314			150		Instrument Test
315-324	2224-2230	R24/23	0, 10, 20, 40 0, 80, 160, 320, 640, 0;		Self broadening
325-335	2145-2148	R1/0	0, 10, 40, 160, 0, 0, 5, 20, 80, 320, 640;		Self broadening
336-342	2135-2145	P1/2	0, 5, 10, 20, 40, 80, 160;		Self broadening
343-349	2058-2065	P19/20	0, 10, 20, 40, 80, 160, 320;		Self broadening
447-450	2224-2230	R24/23	0, 10, 40, 80;		Self broadening
451-452	2224-2230	R24/23	80---	80, 160	Nitrogen broadened
453-454	2224-2230	R24/23	0, 800;		Self broadening
455-466	2145-2148	R1/0	0, 0, 0, 10, 40, 600, 0, 160, 10, 10, 700, 0;		Self broadening
467-469	2145-2148	R1/0	10---	19, 40, 161	Nitrogen broadened
470-471	2145-2148	R1/0	700, 10		Self broadening
472-478	2145-2148	R1/0	10---	10, 20, 40, 81, 161, 323, 242;	Nitrogen broadened
479-481	2145-2148	R1/0	0, 10, 700;		Self broadening
482-487	2180-2185	R10	2.5, 5, 10, 40 160, 640;		Self broadening
488	2180-2185	R10	10---	10	Nitrogen broadened

Scan No.	Wave Number cm <sup>-1</sup>	Lines C <sup>12</sup> O <sup>16</sup>	Pressure mm. Hg.		Remarks
			CO	N <sub>2</sub>	
39-496	2180-2185	R10	5---	10, 20, 40, 80, 161, 323, 646, 646	Nitrogen broadened
97-500	2180-2185	R10	8, 648, 648, 8;		Self broadening
01-507	2180-2185	R10	8---	12, 20, 40, 80, 162, 322, 643;	Nitrogen broadened
08	2180-2185	R10	650		Self broadening
09-516	2145-2150	R1/0	5---	0, 10, 20, 40, 81, 161, 321, 644	Nitrogen broadened
17-524	2213-2217	R20	10, 20, 40, 80, 160, 320, 640, 10		Self broadening
25-531	2213-2217	R20	10---	10, 40, 81, 162, 323, 645, 20;	Nitrogen broadened
32	2213-2217	R20	700;		Self broadening
51-554	2058-2065	P19/P20	80, 750, 0, 750;		Self broadening
55	2058-2065	P19/P20	10---	642	Nitrogen broadened
56	2058-2065	P19/P20	750		Self broadening
57	2135-2150	R1/P2	4	647	Nitrogen broadened
58-561	2135-2150	R1/P2	750		
37-597	2090-2095	P12	20, 5, 10, 20 40, 80, 160, 320, 640, 0, 0;		Self broadening
98-606	2090-2095	P12	1.5---	0, 3, 7, 20, 39, 79, 158, 315, 644;	Nitrogen broadened
07-613	2090-2095	P12	2.5, 5, 10, 20, 40, 80, 640;		Self broadening

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