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OPTOACOUSTIC SPECTROSCOPY OF C₂H₄ AT THE 9 MICROMETERS AND 10 M--ETC(U)
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**OPTOACOUSTIC SPECTROSCOPY OF C_2H_4
AT THE $9\mu M$ AND $10\mu M$
 $C^{12}O_2^{16}$ LASER WAVELENGTHS**

MARCH 1981

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20. ABSTRACT (cont)

absorption were 38.1 percent at the 10 μ m R(20) line, 4.4 percent at the 10 μ m P(18) line, and 18.5 percent at the 10 μ m P(26) laser line.

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1. INTRODUCTION

Ethylene is an atmospheric pollutant produced primarily by auto emissions and other industrial processes using hydrocarbon-based fuels. Concentration levels of 100 ppm are found in uncontrolled gasoline engine exhausts, and similar concentrations have been measured in diesel engine emissions.¹ Ethylene comprises approximately 19.0 percent of the exhausted auto hydrocarbons and 17.6 percent of the photochemical reactivity of these emissions.² Murray and van der Laan found average ethylene concentrations ranging from 2 ppb to over 20 ppb in Palo Alto, and other researchers have reported much higher urban area average concentrations.³

The presence of ethylene concentrations in the environment has two primary effects. First, concentrations of less than 50 ppb can have harmful effects on plant life,³⁻⁶ but are also useful as a ripening agent for commercial fruits.⁵⁻⁷ A second effect is the production of photochemical smog in areas of relatively high hydrocarbon emissions from gasoline engines.²

With the increasing use of CO₂ lasers, the presence of ethylene in a localized environment offers both opportunities and problems to scientists and users of CO₂ lasers. Because of strong absorption at several CO₂ laser transitions, monitoring of ethylene concentration in the atmosphere by using optoacoustic

¹A. C. Stern, Editor, 1966, "Mobile Combustion Sources," Air Pollution, III:62, 68, 84, Academic Press, New York

²A. C. Stern, Editor, 1966, "Effects of Air Pollutants on Vegetation," Air Pollution, I:418, Academic Press, New York

³E. R. Murray and J. E. van der Laan, 1978, "Remote Measurement of Ethylene Using a CO₂ Differential Absorption Lidar," Applied Optics, 17:814

⁴F. B. Abeles and H. E. Heggstad, 1973, "Ethylene: An Urban Air Pollutant," Journal Air Pollution Control, Association, 23:517

⁵S. P. Burg and E. A. Burg, 1965, "Ethylene Action and the Ripening of Foods," Science, 148:1190

⁶D. J. Osborne, 1977, "Ethylene Target Cells in the Growth of Plants," Science Progress, 64:51

⁷P. Perlmutter, S. Shtrikman, and M. Slatkine, 1979, "Optacoustic Detection of Ethylene in the Presence of Interfering Gases," Applied Optics, 18:2267-74

$C^{12}O_2$ ¹⁶ measurement systems has been rapidly developing.^{3,7,8} On the other hand, the presence of unwanted ethylene concentrations in the environment could adversely affect the use of carbon dioxide lasers for various applications such as range finding or communications. Ethylene absorption is relatively strong at wavelengths of untuned CO_2 laser operation; therefore, a precise understanding of the ethylene absorption coefficients at all CO_2 laser wavelengths is important.

Perlmutter et al⁷ have completed an intensive study of optoacoustic detection (based on CO_2 laser instrumentation technology) of ethylene in realistic atmospheric environments. A major concern of the study was the insufficient accuracy of ethylene absorption coefficients; discrepancies larger than 10 percent between various research measurements were quite common. Even though the primary objective was to identify a spectral signature at key wavelengths, repetition of previous optoacoustic measurements of ethylene was considered necessary. In addition, spectral measurements at varying pressures were taken in an attempt to define another mechanism for identifying ethylene in a realistic atmosphere.

CO_2 laser techniques have been used in a number of efforts made to develop accurate ethylene absorption coefficients. The first thorough measurement of ethylene using a CO_2 laser was made by Patty et al and reported in 1974.⁸ That measurement has served as a benchmark for subsequent efforts. Schnell and Fischer reported measurements in 1975,⁹ and Perlmutter et al reported additional unpublished results of Schnell and Fischer in 1979.⁷ In 1977 Shtrikman and Slatkine reported on the development of an intracavity optoacoustic cell (within a CO_2 laser cavity) and its impact on ethylene concentration sensitivity to measurements.¹⁰ Hotta et al¹¹ measured relative absorption for several CO_2 laser lines by ethylene, and Perlmutter et al reported their results in 1979.⁷ In 1978 Mayer et al reported results for

³E. R. Murray and J. E. van der Laan, 1978, "Remote Measurement of Ethylene Using a CO_2 Differential Absorption Lidar," Applied Optics, 17:814

⁷P. Perlmutter, S. Shtrikman, and M. Slatkine, 1979, "Optoacoustic Detection of Ethylene in the Presence of Interfering Gases," Applied Optics, 18:2267-74

⁸R. R. Patty et al, 1974, " CO_2 Laser Absorption Coefficients for Determining Ambient Levels of O_3 , NH_3 , and C_2H_4 ," Applied Optics, 13:2850

⁹P. Perlmutter, S. Shtrikman, and M. Slatkine, 1979, "Optoacoustic Detection of Ethylene in the Presence of Interfering Gases," Applied Optics, 18:2267-74

⁹W. Schnell and G. Fischer, 1975, "Carbon Dioxide Laser Absorption Coefficients of Various Air Pollutants," Applied Optics, 14:2058

¹⁰S. Shtrikman and M. Slatkine, 1977, "Trace-Gas Analysis With a Resonant Optoacoustic Cell Operating Inside the Cavity of a CO_2 Laser," Applied Physics letter, 31:830

¹¹K. Hotta, K. Inoue, and W. Washior, 1978, "Optoacoustic Detection of Ethylene in Exhaust Gas Using a Line-Selectable CO_2 Waveguide Laser," Nippon Electric Company, Kawasaki, Japan, CLEOS 1978 paper THAA4

using an outdoor long-path technique obtained from measuring ethylene absorption.¹² The most recently published absorption coefficients for ethylene were published in May 1980 by Persson et al.¹³ Persson's measurements were also made by using an optoacoustic cell technique similar to that used in our research.

Perlmutter's results reinforce the observation that significant agreement on ethylene absorption coefficients remains elusive, especially at lower absorbing CO₂ laser lines. Table 1 summarizes the percentage difference between Perlmutter's observations and others made to date for the absorption coefficients of ethylene at relatively strong CO₂ absorption lines in the 00°1 to 10°0 CO₂ band. Since the intent was to identify a characteristic spectral signature for ethylene, the weaker 9μm region was ignored, as were numerous other 10μm absorption lines. The results indicate generally increasing measurement discrepancies at wavelengths furthest from the strong P(14) line and weakest in absorptivity of CO₂ laser frequencies. For spectral signature purposes, these results may be adequate; but for accuracy of attenuation calculations at the lesser absorbing CO₂ frequencies in an ethylene environment, the discrepancies can produce significant errors.

In addition to more precise measures of absolute values for the ethylene absorption coefficients in the CO₂ laser frequency range, reliable knowledge of the actual absorption spectra for ethylene will be useful to both researchers and users of CO₂ lasers in ethylene environments. Additionally, molecular structure data (that is, absorption line frequencies, half-widths, and intensities) would assist both groups in the theory and practice relating to CO₂ laser usage. These measurements are becoming increasingly important as the development of laser technology continues.¹⁴

The objectives for the research reported here were to:

- a. Provide a thorough C¹²O₂¹⁶ laser survey of absorption coefficients in the 9μm and 10μm wavelength regions for ethylene as a trace gas at STP;
- b. Compare and analyze the results with previous measurements, especially in the 10μm region;
- c. Review a technique using pressure dependency of absorption at fixed laser probe frequencies and fourier transform spectrometer (FTS) data of moderate resolution for identifying precise absorption line frequencies, half-widths, and intensities;

¹²A. Mayer et al, 1978, "Absorption Coefficients of Various Pollutant Gases at CO₂ Laser Wavelengths; Application to the Remote Sensing of Those Pollutants," Applied Optics, 17:391

¹³J. Persson et al, 1980, "Temperature and Pressure Dependence of NH₃ and C₂H₄ Absorption Cross Sections at CO₂ Laser Wavelengths," Applied Optics, 19:1711

¹⁴E. D. Hinkley, Editor, 1976, "Laser Monitoring of the Atmosphere," Chapter 3, Springer-Verlag, NY

TABLE 1. DISCREPANCIES OF PREVIOUS MEASUREMENTS OF ETHYLENE RELATIVE ABSORPTION COEFFICIENTS IN THE 10 μ m RANGE.^a

Line	Wavelength (μ m)	$\alpha(\lambda)/\alpha(\lambda = 10.5326\mu\text{m})$		Percent Difference from Perlmutter			
		Perlmutter ^b	Patty ^c	Hotta ^d	Mayer ^e	Persson ^f	
R(20)	10.243	0.0421	+3.6%	+8.1%	+113.8%	-12.1%	
P(14)	10.532	1	0	0	0	0	
P(16)	10.548	0.157	-0.6	+6.4	+8.3	+5.1	
P(18)	10.568	0.097	+15.5	+7.2	+8.2	+8.2	
P(20)	10.588	0.0549	-3.7	+10.1	+1.9	+13.0	
P(24)	10.629	0.0357	+24.9	+84.9	+98.9	+107.3	

^a Adapted from table V, Perlmutter, Ref 7

^b Reference 7

^c Reference 8

^d Reference 11

^e Reference 12

^f Reference 13

^g Apparent decimal error in Perlmutter's tabulation

d. Identify additional research needs relative to ethylene absorption coefficients in the 9 μ m and 10 μ m regions.

This report presents results obtained by using a spectrophone as the absorption cell at C¹²O₂¹⁶ laser wavelengths. Values for the absorption coefficients were studied for laser lines from R(4) to R(38) and P(6) to P(42) in the 00⁰1 to 02⁰0 band centered at 9.4 μ m, and from R(4) to R(40) and P(4) to P(40) in the 00⁰1 - 10⁰0 band centered at 10.4 μ m. Studies were also made on the dependence of the absorption on the total pressure, at constant mixing ratio of ethylene to the nitrogen buffer gas, for the lines in the 10P series. Such information can be used in identifying additional research needs relative to ethylene absorption coefficients in the 9 μ m and 10 μ m regions.

A brief overview of the experimental procedure for the survey is included here. (For further details the reader is directed to Brewer and Bruce.¹⁵) The laser radiation was chopped at 920 Hz and passed along the axis of an acoustically isolated resonant subcavity spectrophone^{16,17} with barium fluoride windows. The spectrophone signal was processed by a phase-lock amplifier preceded by a 48 dB per octave bandpass filter to reduce dynamic overload. The ethylene was buffered with "ultrapure" nitrogen to a total pressure of 760 torr at 295°K and was introduced into the spectrophone. The concentrations used varied from 20 to 1600 ppm. One or two lines were monitored during each spectral run to give correction factors for any temporal changes in concentration (at most a few percent per day). In taking the pressure dependence data, the spectrophone was simply pumped down to the desired pressures, keeping the relative ethylene to nitrogen concentration constant.

Each set of spectral data represented a nearly complete pattern of relative absorption coefficients for ethylene. Each data set was scaled against the other sets in an iterative fashion to produce the final pattern. This final pattern was then scaled to absolute values by comparison of the higher absorption values with those obtained by Patty⁸ who used the White cell method.

¹⁵R. J. Brewer and C. W. Bruce, 1978, "Photoacoustic Spectroscopy of NH₃ at the 9 μ m and 10 μ m C¹²O₂¹⁶," Applied Optics, 17:3746

¹⁶C. W. Bruce et al, 1976, "Application of Pulsed-Source Spectrophone to Absorption by Methane at DF Laser Wavelengths," Applied Optics, 15:2970

¹⁷C. W. Bruce and R. G. Pinnick, 1977, "In-Situ Measurements of Aerosol Absorption With a Resonant CW Laser Spectrophone," Applied Optics, 16:1762

⁸R. R. Patty et al, 1974, "CO₂ Laser Absorption Coefficients for Determining Ambient Levels of O₃, NH₃, and C₂H₄," Applied Optics, 13:2850

Figures 1 through 4 provide a comparison of typical spectrophotometer traces (resolution $\approx 1 \text{ cm}^{-1}$) of C_2H_4 absorption coefficients with the spectrophone measurements made in this study. The figures show that the magnitudes of the variations between the high and low resolution measurements are greatest (nearly an order of magnitude in some instances) in wavelength regions of weakest absorption.

Table 2 presents the high-resolution absorption coefficients of ethylene at the $9\mu\text{m}$ and $10\mu\text{m}$ CO_2 laser lines, along with the probable errors in measurement. The errors due to the absolute calibration, which add as $[(\% \text{ P.E.})^2 + (5.4\%)^2]^{1/2}$, are not included. The absorption due to ethylene at $10\mu\text{m}$ wavelengths is considerably stronger than for absorption at $9\mu\text{m}$ wavelengths, with the strongest absorption occurring at the 10 P laser lines. The wavelengths and frequencies in air were obtained from Chang.^{1*}

Even though Patty's⁸ absorption coefficients were used to calibrate our data, there is a significant variation between the two spectral patterns. Comparing all common data points (R(10) to R(32) and P(10) to P(34) in the $10\mu\text{m}$ range and R(10) to R(32) and P(10) to P(34) in the $9\mu\text{m}$ range), the average mean discrepancy between the two sets of data is 1.14 percent in the $10\mu\text{m}$ range (standard deviation = 22.1 percent) and 29.8 percent in the $9\mu\text{m}$ range (standard deviation = 18.1 percent).

Although the average mean difference is reasonably small in the $10\mu\text{m}$ range (as would be expected of the data used for calibration of our values), the absolute difference between the data of Patty and of ASL is an average mean of 15.9 percent (standard deviation = 15.0 percent). This comparison is provided in table 3.

These results are consistent with Patty's⁸ observation that his White cell technique tends to overestimate the lower values for absorption coefficients since the systematic difference in the $9\mu\text{m}$ range shows Patty's values nearly 30 percent above ours.

The differences between the two spectral patterns suggest problems in the measurement techniques (for example, system purity, linearity, precision) and/or data analysis.

^{1*}T. Y. Chang, 1970, "Accurate Frequencies and Wavelengths of CO_2 Laser Lines," Optical Communication, 2:77

⁸R. R. Patty et al, 1974, " CO_2 Laser Absorption Coefficients for Determining Ambient Levels of O_3 , NH_3 , and C_2H_4 ," Applied Optics, 13:2850

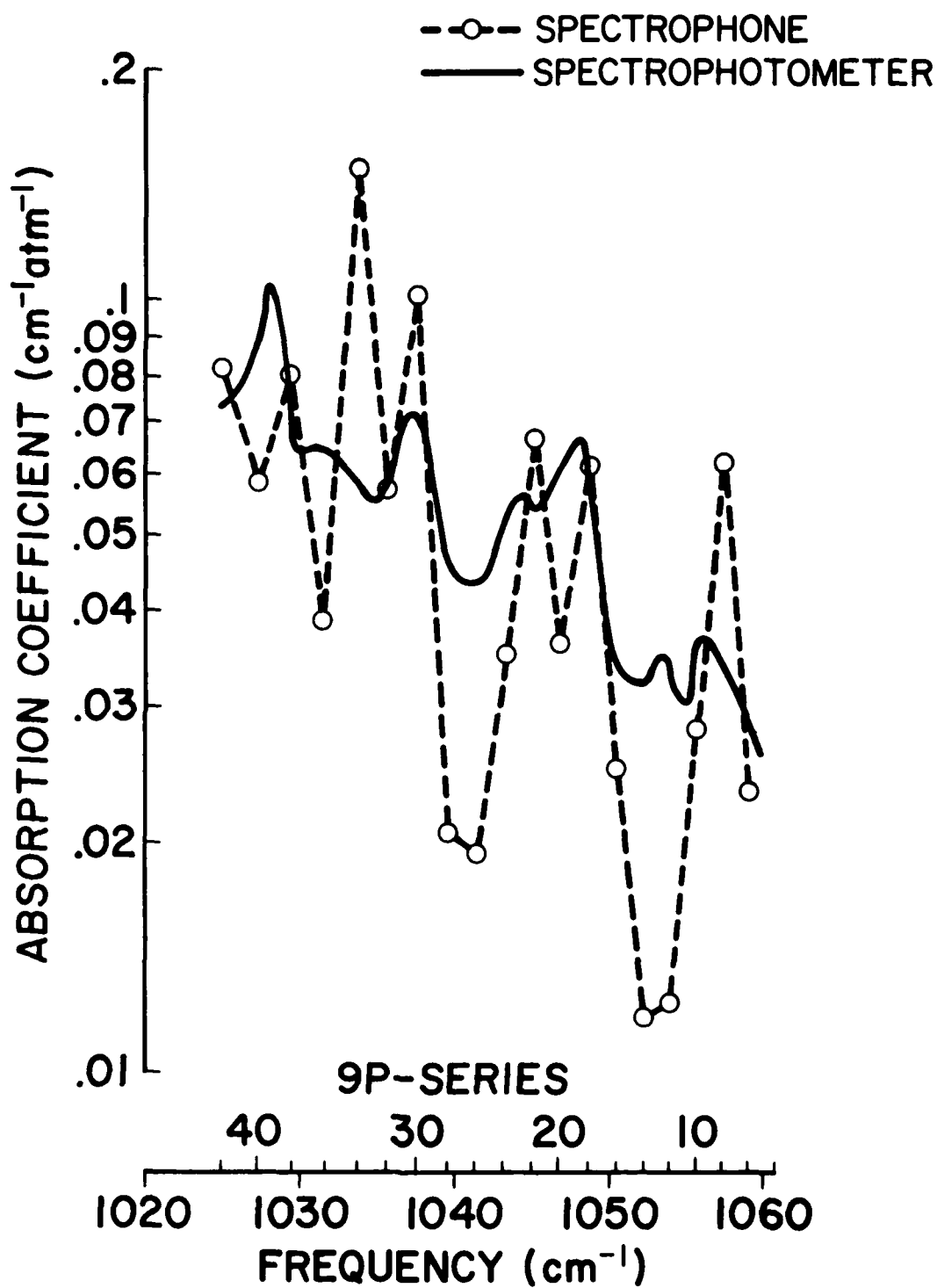


Figure 1. Comparison of spectrophone and spectrophotometer spectra for ethylene absorption in the $10\mu\text{m}$, P-Series.

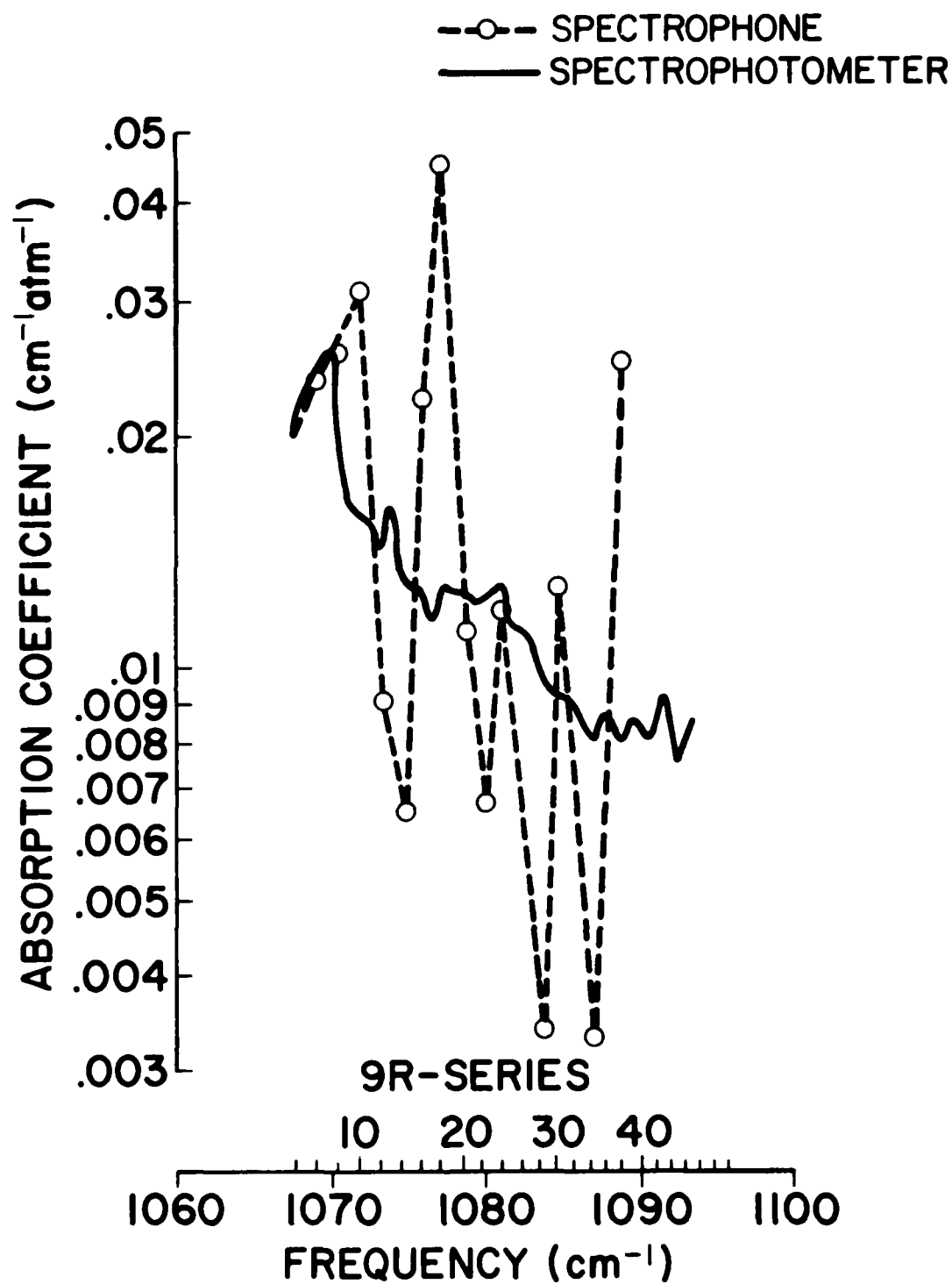


Figure 2. Comparison of spectrophone and spectrophotometer spectra for ethylene absorption in the $10\mu\text{m}$, R-Series.

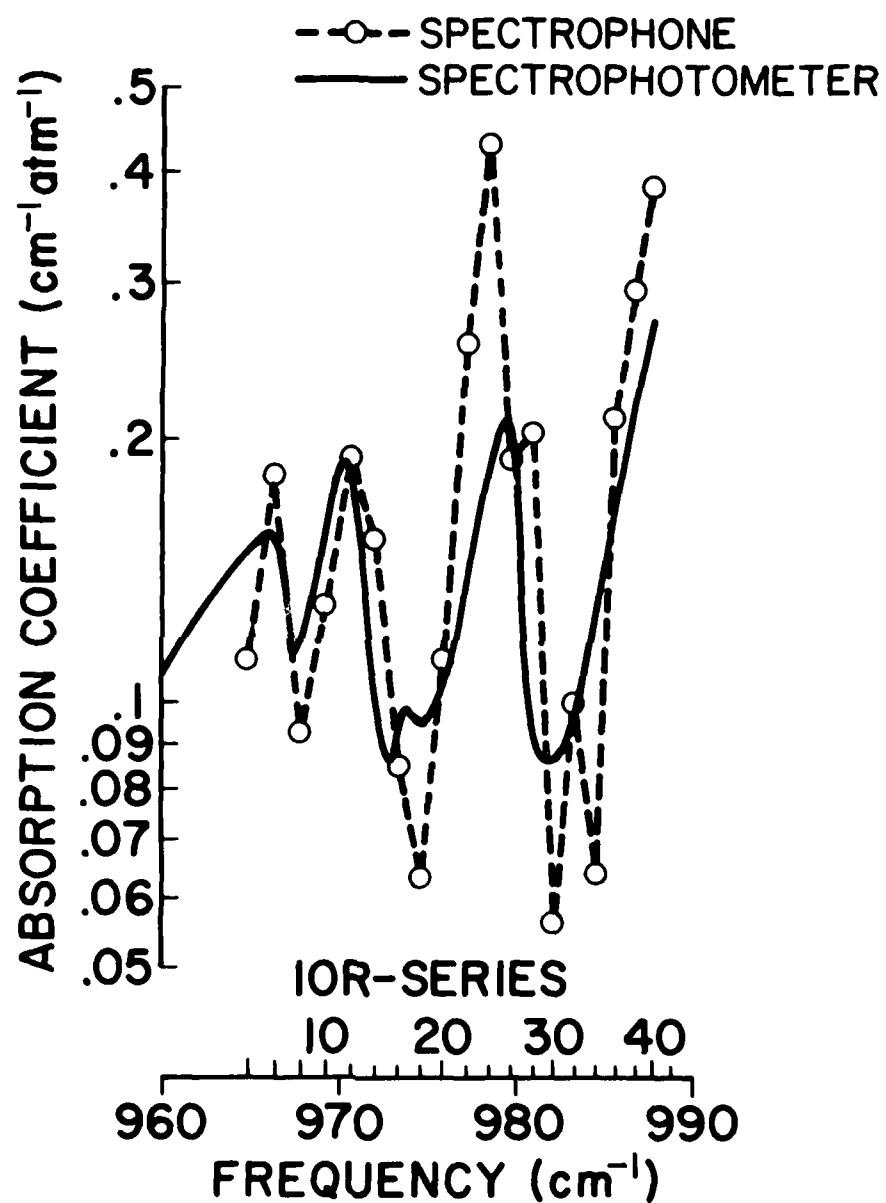


Figure 3. Comparison of spectrophone and spectrophotometer spectra for ethylene absorption in the $9\mu\text{m}$, R-Series.

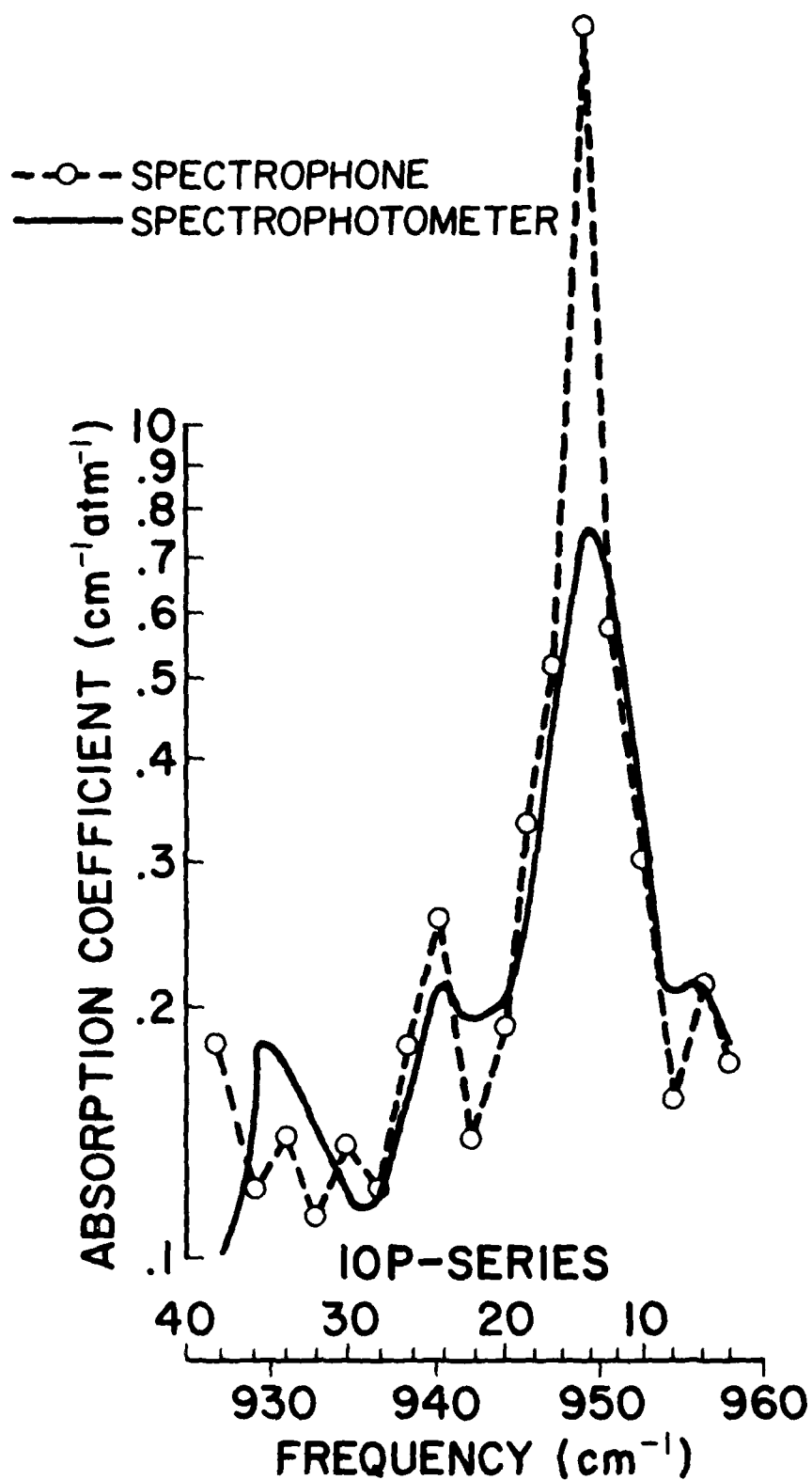


Figure 4. Comparison of spectrophone and spectrophotometer spectra for ethylene absorption in the $9\mu\text{m}$, P-Series. Both traces are the result of ASL efforts.

TABLE 2. ABSORPTION COEFFICIENT OF C_2H_4 STP

Spectral Line	00°1 - 02°0 CO ₂ band				00°1 - 10°0 CO ₂ band			
	Wavelength ^a (air, μ m)	Frequency (cm^{-1})	Coefficient ($atm\text{-}cm$) ⁻¹	Error ^b ($\pm$ %)	Wavelength ^a (μ m air)	Frequency (cm^{-1})	Coefficient ($atm\text{-}cm$) ⁻¹	Error ^b ($\pm$ %)
R(40)	9.1740695	1090.0280	---	---	10.125340	987.62115	3.82	7.4
R(38)	9.1826656	1089.0314	0.0320	3.0	10.136208	986.56822	2.99	7.9
R(36)	9.1916115	1087.9485	0.254	7.8	10.147246	985.48911	2.13	6.4
R(34)	9.2007332	1086.8699	0.0330	7.3	10.158637	984.38399	0.634	5.8
R(32)	9.2100919	1085.7655	0.0455	7.9	10.170322	983.25298	1.02	6.3
R(30)	9.2196897	1084.6352	0.130	7.4	10.182301	982.09624	0.553	6.0
R(28)	9.2295299	1083.4788	0.0342	8.7	10.194575	980.91391	2.02	5.8
R(26)	9.2396143	1082.2962	0.0720	5.8	10.207143	979.70612	1.89	5.8
R(24)	9.2499459	1081.0874	0.119	7.9	10.220006	978.47298	4.33	6.2
R(22)	9.2605266	1079.8522	0.0667	5.4	10.233167	977.21461	2.55	6.0
R(20)	9.2713584	1078.5906	0.115	8.7	10.246625	975.93112	1.09	5.5
R(18)	9.2824439	1077.3025	0.448	6.1	10.260381	974.62263	0.617	7.4
R(16)	9.2851562	1076.9878	0.224	6.6	10.274438	973.28921	0.914	6.4
R(14)	9.3053855	1074.6465	0.0654	5.4	10.288797	971.93094	1.41	5.6
R(12)	9.3172462	1073.2785	0.0904	8.5	10.303459	970.54793	1.66	5.8
R(10)	9.3293703	1071.8837	0.308	6.8	10.318424	969.14023	1.31	5.9
R(8)	9.3417582	1070.4623	0.253	8.3	10.333697	967.70792	0.924	5.6
R(6)	9.3544136	1069.0141	0.238	9.1	10.349277	966.25105	1.31	5.7
R(4)	9.3673384	1067.5391	0.198	5.8	10.365168	964.676967	1.16	6.6
P(4)	9.4288858	1060.5707	---	---	10.40579	957.80124	1.68	5.5
P(6)	9.4433281	1058.9487	0.237	8.7	10.458220	956.18568	2.06	7.7
P(8)	9.4580517	1057.3002	0.626	5.5	10.476187	954.54580	1.52	6.0
P(10)	9.4730501	1055.6251	0.276	5.7	10.494484C	952.88157	2.98	6.4
P(12)	9.4883547	1053.9235	0.124	7.3	10.513117C	951.19299	4.31	6.4
P(14)	9.5039363	1052.1956	0.119	9.8	10.532080C	949.48003	30.4	7.6
P(16)	9.5198085	1050.4413	0.177	6.3	10.551387C	947.74271	5.07	7.2
P(18)	9.5359720	1048.6608	0.607	6.3	10.571037C	945.98096	3.32	6.7
P(20)	9.5524277	1046.8543	0.359	5.9	10.591035	944.19476	1.84	6.2
P(22)	9.5691793	1045.0217	0.661	5.7	10.611385	942.38407	1.37	6.5
P(24)	9.5862176	1043.1633	0.332	6.1	10.632090C	940.54883	2.29	5.8
P(26)	9.6035731	1041.2791	0.193	5.8	10.653156	938.68898	1.78	5.9
P(28)	9.6212193	1039.3693	0.202	7.5	10.674586	936.80447	1.19	6.1
P(30)	9.6391665	1037.4341	1.03	6.5	10.696386	934.89522	1.35	6.2
P(32)	9.6574158	1035.4737	6.563	5.7	10.718560	932.96114	1.10	6.0
P(34)	9.6759701	1033.4881	1.50	5.7	10.741113	931.00217	1.40	6.8
P(36)	9.6953949	1031.4775	0.385	5.9	10.764052	929.01820	1.20	7.7
P(38)	9.7139976	1029.4423	0.791	5.6	10.787380	927.00913	1.82	5.7
P(40)	9.7334732	1027.3825	0.570	8.8	10.811104	924.97486	0.615	6.0
P(42)	9.7532592	1025.2983	0.815	5.4	10.835231	922.91528	---	---

a The wavelengths and frequencies in air were obtained from reference 18.

b See text.

c Absorptions at these lines were used to calibrate the spectrophone.

TABLE 3. COMPARISON OF ETHYLENE (C₂H₄) ABSORPTION COEFFICIENTS FOR ASL AND PATTY^a

00°1 - 02°0 CO₂ band 9μm

Absorption coefficients
(atm-cm)⁻¹

Spectral Line	Wavelength (air, μm)	ASL	Patty ^a	ASL/Patty (percent)	Δα (Pat-ASL)	Δα/ASL (percent)
R(30)	9.217	0.130	0.22	59.1	0.09	69.2
R(28)	9.227	0.0342	0.046	74.4	0.0118	34.5
R(26)	9.237	0.0720	0.11	65.5	0.038	52.8
R(24)	9.247	0.1190	0.18	66.1	0.061	51.3
R(22)	9.258	0.0667	0.10	66.7	0.0333	49.9
R(20)	9.269	0.1150	0.17	67.7	0.0550	47.8
R(18)	9.280	0.448	0.61	73.4	0.1620	36.2
R(16)	9.291	0.224	0.28	80.0	0.0560	25.0
R(14)	9.303	0.0654	0.073	89.6	0.0076	11.6
R(12)	9.315	0.0904	0.082	110.2	-0.0084	-9.3
R(10)	9.327	0.3080	0.33	93.3	0.0220	7.1
P(10)	9.470	0.276	0.42	65.7	0.1440	52.2
P(12)	9.486	0.124	0.16	77.5	0.0360	29.0
P(14)	9.501	0.119	0.14	85.0	0.0210	17.7
P(16)	9.517	0.177	0.20	88.5	0.0230	13.0
P(18)	9.533	0.607	0.79	76.8	0.1830	30.2
P(20)	9.550	0.359	0.40	89.8	0.0410	11.4
P(22)	9.567	0.661	0.82	80.6	0.1590	24.1
P(24)	9.584	0.332	0.42	79.1	0.0880	26.5
P(26)	9.601	0.193	0.23	83.9	0.0370	19.2
P(28)	9.619	0.202	0.24	84.2	0.0380	18.8
P(30)	9.637	1.03	1.27	81.1	0.2400	23.3
P(32)	9.655	0.563	0.72	78.2	0.1570	27.9
P(34)	9.673	1.50	2.18	68.8	0.6800	45.3
		$\bar{x}$		$\bar{x}$	0.0990	29.8
		s		s	0.1400	18.1
		Absolute Values		$\bar{x}$	0.0997	30.6
				s	0.1395	16.7

^aSee reference (8). ASL data taken at STP, Patty data taken at std pressure and 300°K.

TABLE 3 (cont)

00°1 - 10°0 CO₂ band 10 μ m

Absorption coefficients -1 (atm-cm)						
Spectral Line	Wavelength (air, μm)	ASL	Patty ^a	ASL/Pat (percent)	Δα (Pat-ASL)	Δα/ASL (percent)
R(32)	10.168	1.02	0.83	122.9	-0.1900	-18.6
R(30)	10.180	0.553	0.56	98.75	0.007	1.3
R(28)	10.192	2.02	2.35	86.0	0.3300	16.3
R(26)	10.204	1.89	2.16	87.5	0.2700	14.3
R(24)	10.217	4.33	5.04	85.9	0.7100	16.4
R(22)	10.230	2.55	2.64	96.6	0.0900	3.5
R(20)	10.244	1.09	1.27	85.8	0.1800	16.5
R(18)	10.258	0.617	0.55	112.2	-0.0670	-10.9
R(16)	10.272	0.914	1.04	87.9	0.1260	13.8
R(14)	10.286	1.41	1.27	111.0	-0.1400	-9.93
R(12)	10.301	1.66	1.94	85.6	0.2800	16.9
R(10)	10.316	1.31	1.51	86.8	0.2000	15.3
P(10)	10.492	2.98	3.10	96.1	0.1200	4.03
P(12)	10.510	4.31	4.35	99.1	0.0400	0.9
P(14)	10.529	30.4	29.10	104.5	-1.3000	-4.3
P(16)	10.549	5.07	4.55	111.4	-0.5200	-10.3
P(18)	10.568	3.32	3.28	101.2	-0.0400	-1.2
P(20)	10.588	1.84	1.64	112.2	-0.2000	-10.87
P(22)	10.603	1.37	1.09	127.5	-0.2800	-20.44
P(24)	10.629	2.29	2.10	109.1	-0.1900	-8.30
P(26)	10.650	1.78	2.40	74.2	0.6200	34.83
P(28)	10.672	1.19	1.30	91.5	0.1100	9.24
P(30)	10.693	1.35	1.63	82.8	0.2800	20.74
P(32)	10.716	1.10	0.48	229.2	-0.6200	-56.36
P(34)	10.738	1.40	0.54	259.3	-0.8600	-61.43
				$\bar{x}$	-0.0418	-1.14
				s	0.4382	22.1
Absolute Values				$\bar{x}$	0.3108	15.9
				s	0.3053	15.0

Table 4 provides a second comparison with existing data. The data are based on Perlmutter's presentation,⁷ and his data are compared with the data of ASL, Hotta,¹¹ Patty,⁸ Mayer,¹² and Persson.¹³ The results presented in table 4 differ importantly from those discussed above in that they compare relative rather than absolute values; the 10P(14) line acts as the normalizing value. The discrepancies between the four sets for each probe line are indicated in the ratio of the standard deviation to the mean absorption coefficient s/x .

These discrepancies range from 4.4 to 38.1 percent. While the P(18) and P(20) cases are in substantial agreement, those remaining have a greater than 10 percent s/x ratio and differ significantly from one another.

Note that the values provided in table 4 are derived from six distinct experimental techniques. Four techniques--ASL, Perlmutter's, Hotta's, and Persson's--used optoacoustic methods varying from one another, while Patty used the White cell methodology and Mayer used an outdoor long-path technique.

Kelley et al.¹⁹ found that discrepancies of up to 30 percent existed between various measurements and calculations of CO_2 absorption coefficients (of various atmospheric components and air itself). The explanation offered focused on the lack of experimental observations, the error inherent in those existing at that time, and the temperature dependence of line strengths (2.2 percent per $^\circ\text{K}$).

In the comparison of table 4, Kelley's explanations of the discrepancies do not appear to be applicable. First, the experimental uncertainty referred to is related to long-path measurement techniques (White cell), and only two of the data sets here were taken with this technique. Secondly, although there is apparent temperature variation between Patty's data and the other data,

⁷P. Perlmutter, S. Shtrikman, and M. Slatkine, 1979, "Optoacoustic Detection of Ethylene in the Presence of Interfering Gases," Applied Optics, 18:2267-74

¹¹K. Hotta, K. Inoue, and W. Washio, 1978, "Optoacoustic Detection of Ethylene in Exhaust Gas Using a Line-Selectable CO_2 Waveguide Laser," Nippon Electric Company, Kawasaki, Japan, CLEOS 1978 paper THAA4

⁸R. R. Patty et al, 1974, " CO_2 Laser Absorption Coefficients for Determining Ambient Levels of O_3 , NH_3 , and C_2H_6 ," Applied Optics, 13:2850

¹²A. Mayer et al, 1978, "Absorption Coefficients of Various Pollutant Gases at CO_2 Laser Wavelengths; Application to the Remote Sensing of Those Pollutants," Applied Optics, 17:391

¹³U. Persson et al, 1980, "Temperature and Pressure Dependence of NH_3 and C_2H_6 Absorption Cross Sections at CO_2 Laser Wavelengths," Applied Optics, 19:1711

¹⁹P. L. Kelley and R. A. McClatchey, 1976, "Molecular Absorption of Infrared Laser Radiation in the Natural Atmosphere," Optical Quantum Electron, 8:117

TABLE 4. RELATIVE SPECTRAL SIGNATURES OF ETHYLENE AT STPa

$$\frac{\epsilon(\lambda)}{\epsilon(\lambda = 10.5326 \mu\text{m})}$$

Line	Wavelength (μ m)	ASL	Perlmutter ^b	Patty ^c	Hotta ^d	Mayer ^e	Persson ^f	X	S	S/X (%)
R(20)	10.243	0.036	0.042	0.044	0.045	0.090	0.037	0.049	0.019	38.1
P(14)	10.532	1	1	1	1	1	1	---	---	---
P(16)	10.548	0.117	0.157	0.156	0.167	0.170	0.165	0.155	0.018	11.5
P(18)	10.568	0.109	0.097	0.112	0.104	0.105	0.105	0.105	0.005	4.4
P(20)	10.588	0.060	0.058 ^g	0.056 ^g	0.064	0.055	0.061	0.059	0.003	5.2
P(24)	10.629	0.075	0.076	0.072	0.053	0.071	0.074	0.070	0.008	11.2
P(26)	10.650	0.059	0.036	0.045	0.066	0.060	0.060	0.054	0.010	18.5

^a Based on Perlmutter table V (Ref 7)^b Reference (7)^c Reference (8)^d Reference (11)^e Reference (12)^f Reference (13)^g Apparent decimal error in Perlmutter's tabulation

this variation should produce a relatively consistent bias in his data versus the other. However, this consistency does not appear to be the case since the discrepancies are generally erratic. Discrepancies could be due to impurities--particularly if the values are quite repeatable as in our case. These discrepancies were found to be highly likely in a previous comparison of White cell and spectrophone measurements.¹⁶ In that case the authors' spectrophone system agreed well with several White cell results but disagreed with other published spectrophone results. For the present paper, the high purity gas was handled in a largely stainless steel, highly purified gas mixing system previously described.¹⁵ An analysis using a mass spectrometer integral to this system was inconclusive because of interfering mass numbers.

2. PRESSURE DEPENDENCY

Center frequencies of absorption lines relative to those of laser probe lines may be inferred from data on absorption as a function of total pressure. Conversely, Perlmutter et al⁷ suggest that observed pressure dependency of ethylene absorption coefficients could serve to help identify ethylene concentrations in the atmosphere through "pressure tuning" a spectrophone for a small number of laser probe lines. Perlmutter et al provided limited data on pressure dependency for ethylene absorption coefficients in the 10 μ m region and appear to be the only authors to do so to date.

The results of our pressure dependence measurements are graphically illustrated in figure 5. The curves represent the best-fit approximations of the relationship between the total spectrophone pressure, P, and the relative absorption coefficient for the (6) through the (24) CO₂ laser transition lines in the 10 μ m band. It is unfortunate that the data provided by Perlmutter et al⁷ are not readily comparable with our experimentally derived pressure-absorption relationships. (In addition, there are inconsistencies in their pressure data which make the validity of any possible comparisons questionable.*)

¹⁶C. W. Bruce et al, 1976, "Application of Pulsed-Source Spectrophone to Absorption by Methane at DF Laser Wavelengths," Applied Optics, 15:2970

¹⁵R. J. Brewer and C. W. Bruce, 1978, "Photoacoustic Spectroscopy of NH₃ at the 9 μ m and 10 μ m C¹²O₂¹⁶ Laser Wavelengths," Applied Optics, 17:3746

⁷P. Perlmutter, S. Shtrikman, and M. Slatkine, 1979, "Optoacoustic Detection of Ethylene in the Presence of Interfering Gases," Applied Optics, 18:2267-74

*In particular Perlmutter's relative α for 10P(26) is given as 0.0357 in his table V and 0.08 in his table VI. The first α (λ) is for STP and the second at presumably the same temperature and partial pressure for C₂H₄, but at 1000 mbar vs 1013 mbar (standard). In addition, in his table VI, the data for the 10R(24) transition are mislabelled P(24), including the correct wavelength for the 10P(24) transition. Further, the data presented in table VI are inconsistent for some values with data presented in the previous tables.

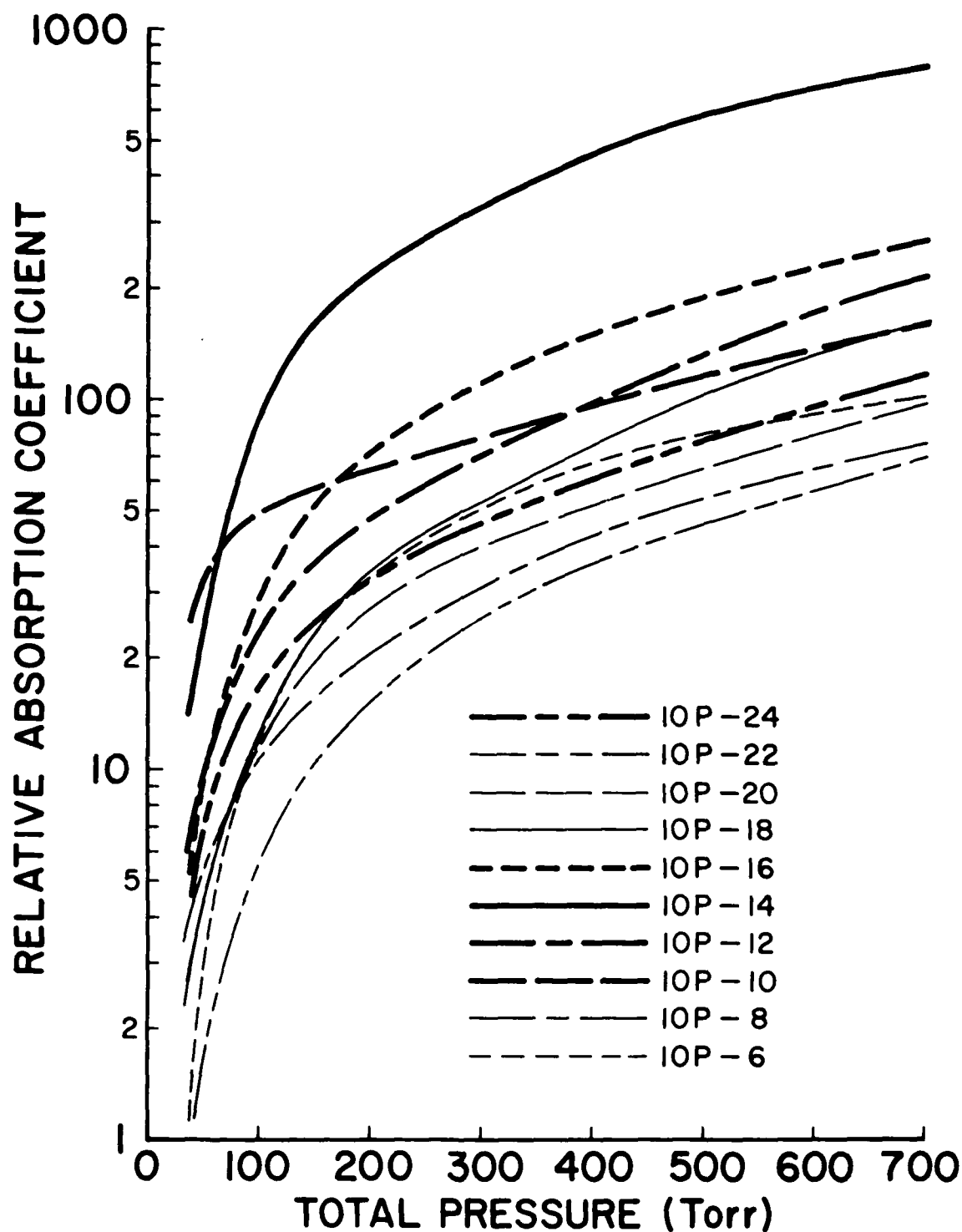


Figure 5. Dependence of the 10μm P-Band $C^{12}O_2^{16}$ laser absorption coefficients in ethylene on total pressure. Data taken for concentrations of C_2H_4 in N_2 at approximately 24, 72, and 240 ppm standard temperature. The curves represent averages for a number of pressure dependent measurements. Standard deviation of the values up to 6 percent of the relative value.

Perlmutter also provides data at 360 torr and compares relative spectral signatures with previously unpublished data. Table 5 compares our data with the data provided by Perlmutter.⁷ Since our data were interpolated from experimental observations at 295 and 395 torr (rather than direct at 360 torr), the consistently higher values in our data could be due to the interpolation on a nonlinear curve. While a maximum discrepancy of 25 percent (10P(20)) exists between our data and Perlmutter's, the general patterns are similar; and as we have already observed, this magnitude of experimental disagreement is typical in measuring relative absorption coefficients of ethylene.

If we assume that the pressure dependence for an absorbing gas is described by a Lorentz shaped line over the range of pressures studied, then for a total pressure P, the absorption coefficient α varies as

$$\sum_i \frac{S_i \gamma_i P}{(\nu - \nu_i)^2 + \gamma_i^2} \quad (1)$$

where S_i , γ_i , and ν_i , are the intensity, width at half maximum, and position of the i^{th} line contributing to the attenuation of the radiation; and ν is the probe frequency. The width γ_i may be written $\gamma_i = \sum_j \gamma_{ij} P_j$ to display the broadening contributions of the constituent gases (j), including the "self" broadening term. If the mixing ratio for the absorber to buffer gas is $\ll 1$, and it is held constant over the conditions, then the absorption coefficient varies as

$$\alpha \approx \sum_i \frac{S_i}{\frac{(\nu - \nu_i)^2 + 1}{\gamma_i P}} \quad (2)$$

Expression (2) shows that α is particularly sensitive to pressure changes in the transition region $\nu - \nu_i \approx \gamma_i P$.

Thus a potential application of the relationship between absorption coefficient and pressure is the high resolution identification of absorbing line center frequencies. The bases for this application are the sensitivity of the relationship in the transition region and the existence of numerous fixed and well-known laser line frequencies. This technique is merely supportive of other spectroscopic data since line shapes (best obtained by using tunable diodes--absolute frequencies not required), strengths, and

⁷P. Perlmutter, S. Shtrikman, and M. Slatkine, 1979, "Optoacoustic Detection of Ethylene in the Presence of Interfering Gases," Applied Optics, 18:2267-74

TABLE 5. COMPARISON OF C₂H₄ ABSORPTION COEFFICIENTS FROM THREE SOURCES
AT 360 TORR PRESSURE

Line	Wavelength (μ m)	ASL ^a	Perlmutter ^b	Schnell ^c
10P(14)	10.532	1	1	1
10P(16)	10.548	0.19	0.16	0.17
10P(18)	10.568	0.094	0.084	0.089
10P(20)	10.588	0.068	0.054	0.063

^a Interpolated from 395 and 295 torr observations

^b Reference (7)

^c Unpublished data (see reference 7)

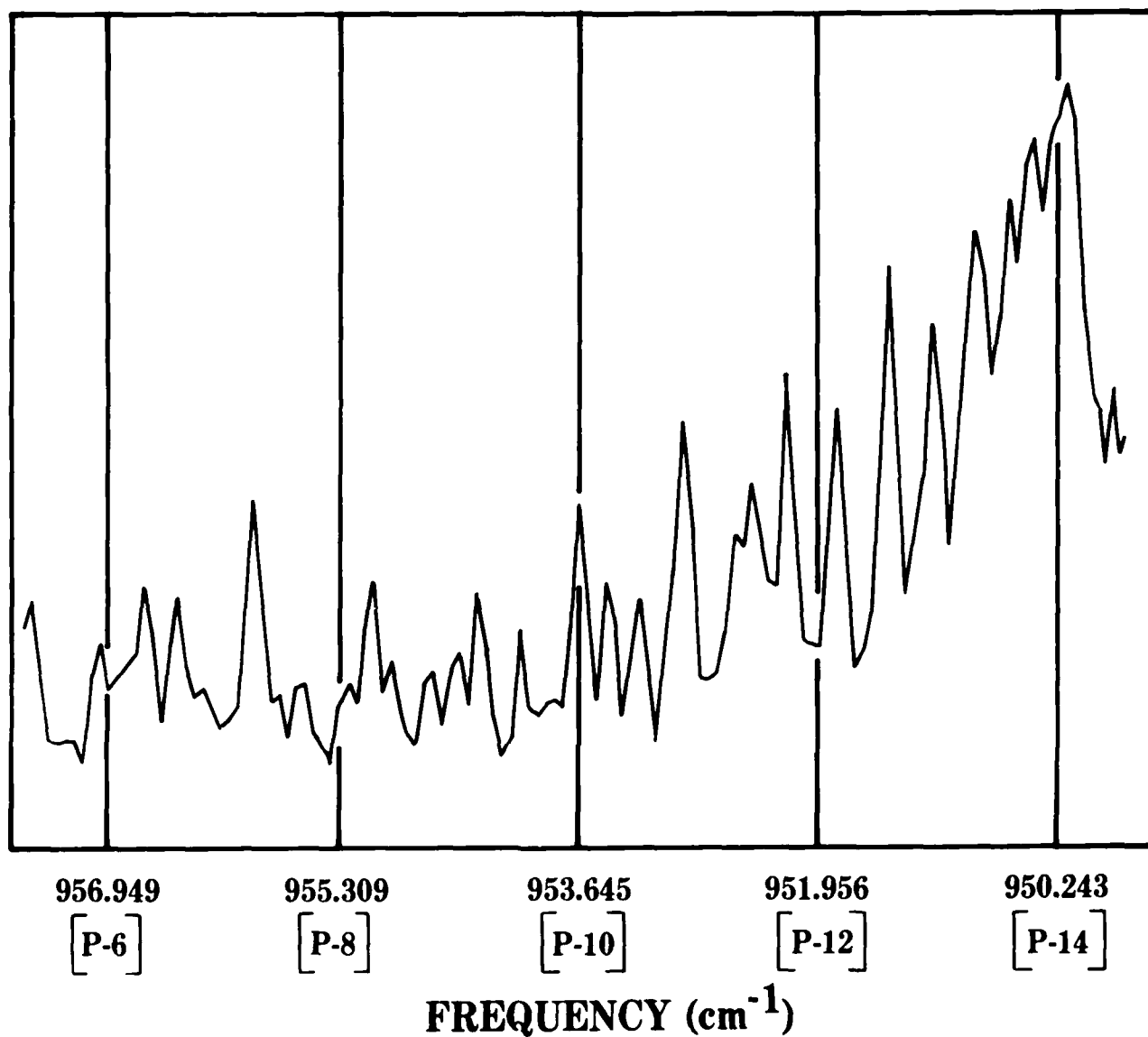
approximate position (using instruments of lower resolution) must be known. If the underlying structure is sufficiently complex, the process will be ambiguous. An enhancement in resolution also depends on the existence of nearby laser probe lines. With regard to the latter point, two additional isotopes of CO_2 are available for use in laser systems, greatly increasing the number of probe frequencies.

To demonstrate the potential for the pressure dependency technique in absorption frequency identification, we have examined two CO_2 laser lines ($10\mu\text{m P}(10)$, $10\mu\text{m P}(12)$) in depth and developed an estimate of the C_2H_4 molecular absorption frequency near the $10\mu\text{m P}(10)$ CO_2 laser line. The theory described in equations (1) and (2) will be useful only if the absorbing line and the laser probe line are very close. Otherwise, the multiplicity of the absorbing lines will present a relatively smoothly decreasing pressure-to-absorption curve and the distinctions between absorbing lines will be quickly lost. If an absorbing line and a laser line are in close proximity, a more sustained absorption coefficient with pressure decrease will occur until the transition region is reached, providing a better opportunity to identify the absorbing line frequency.

A comparison of the $10\mu\text{m P}(10)$ pressure dependence with the other coefficients of figure 5 suggests that the $10\mu\text{m P}(10)$ must be quite close to a strongly absorbing ethylene line. The $10\mu\text{m P}(12)$ line, for example, has a shape similar to the other curves. Figure 6 is the portion of a high-resolution FTS trace (average of 100 scans) of a high concentration of ethylene at low pressure (about 100 torr). $\text{C}^{12}\text{O}_2^{16}$ laser spectral lines have been carefully plotted on the FTS trace. From this plot, the $10\mu\text{m P}(10)$ line appears to be very close to a strongly absorbing C_2H_4 line, while the $10\mu\text{m P}(12)$ line is in the trough between two strongly absorbing lines. From the trace we would have to conclude that the C_2H_4 line near the $10\mu\text{m P}(10)$ laser line has a slightly shorter wavelength than the laser line itself.

Figure 7 provides a comparison of the theoretical plots of the absorbing C_2H_4 frequency for various $\Delta\nu_i = \nu - \nu_i$ and the experimental observations of the pressure dependency of the $10\mu\text{m P}(10)$ laser line. A best curve fit yields $\Delta\nu = 0.0065 \text{ cm}^{-1}$ which yields a frequency of 953.639 cm^{-1} for the C_2H_4 absorption line.

Figure 8 provides a similar comparison for the absorption coefficient at the $10\mu\text{m P}(12)$ laser line using a calculated Lorentzian single absorption line fit with a $\Delta\nu$ of 0.14 cm^{-1} , the approximate FTS-measured wavelength difference between the $10\mu\text{m P}(12)$ line and the closest C_2H_4 strong absorption line. The difference in absorption profiles suggests that the underlying structure is more complicated than the model in this case. At large values of $\Delta\nu$ we might expect to encounter, as in the case of $10\mu\text{m P}(12)$, weak underlying molecular absorption lines of the trace gas. These lines complicate the analysis at lower total pressures.



CO₂ Laser Line Designation

Figure 6. Comparison of FTS ethylene absorption spectra with selected $C^{12}O_2^{16}$ laser transitions in the 10 μ m P-Series. C_2H_4 concentrations were roughly 5 parts per thousand in N_2 . FTS total pressure was at approximately 120 torr, standard temperature. The FTS was calibrated by using known absorption lines elsewhere in the spectrum, and the placement of the laser transition lines is estimated at 0.001 cm^{-1} .

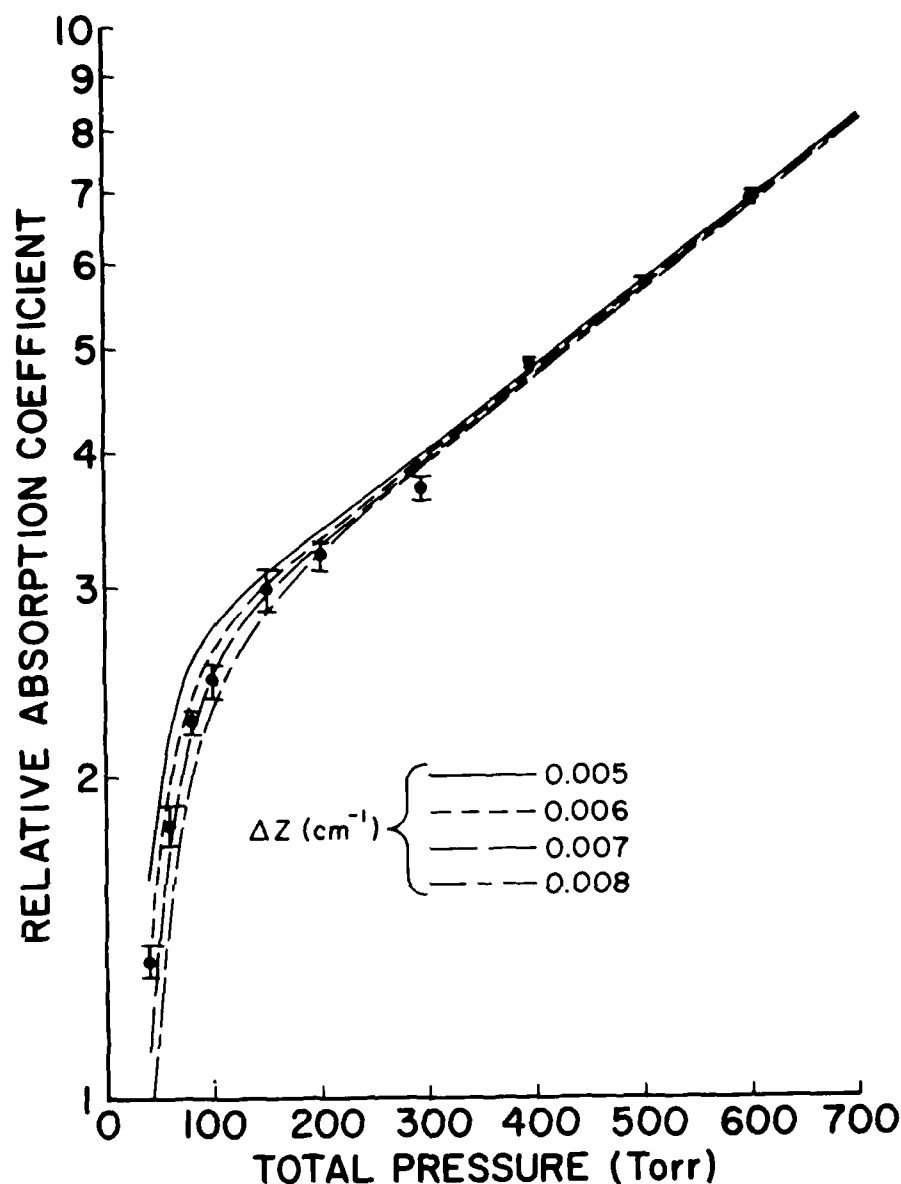


Figure 7. Experimental versus theoretical pressure dependence for the C_2H_4 absorption line near the $10P(10) C^{12}O_2^{16}$ laser line. To develop a close fit to the experimental data, $\alpha(\nu)$ was considered equal to the sum

$$\left(\frac{S_1 \gamma_1}{\Delta \nu_1^2} + \frac{S_2 \gamma_2}{\Delta \nu_2^2} + \frac{S_3 \gamma_3}{\Delta \nu_3^2} \right),$$

where $\Delta \nu_i$ represents the distance between the absorption line ν_i and the laser line frequency ν . $\Delta \nu_1$ was the absorption line of interest (closest to the $10P(10)$ line) and the values of $\alpha(\nu)$ (relative) for various $\Delta \nu_i$ are graphed in the figure. Line strength S_i was taken as unity. $\Delta \nu_2$ was the next nearest line at 0.22 cm^{-1} and $S_2 = 0.75$ and $\Delta \nu_3$ represented all other contributing absorption lines at $\Delta \nu_3 = 0.50$ and $S_3 = 2.9$. S_1 was initially varied until a good curve fit was achieved.

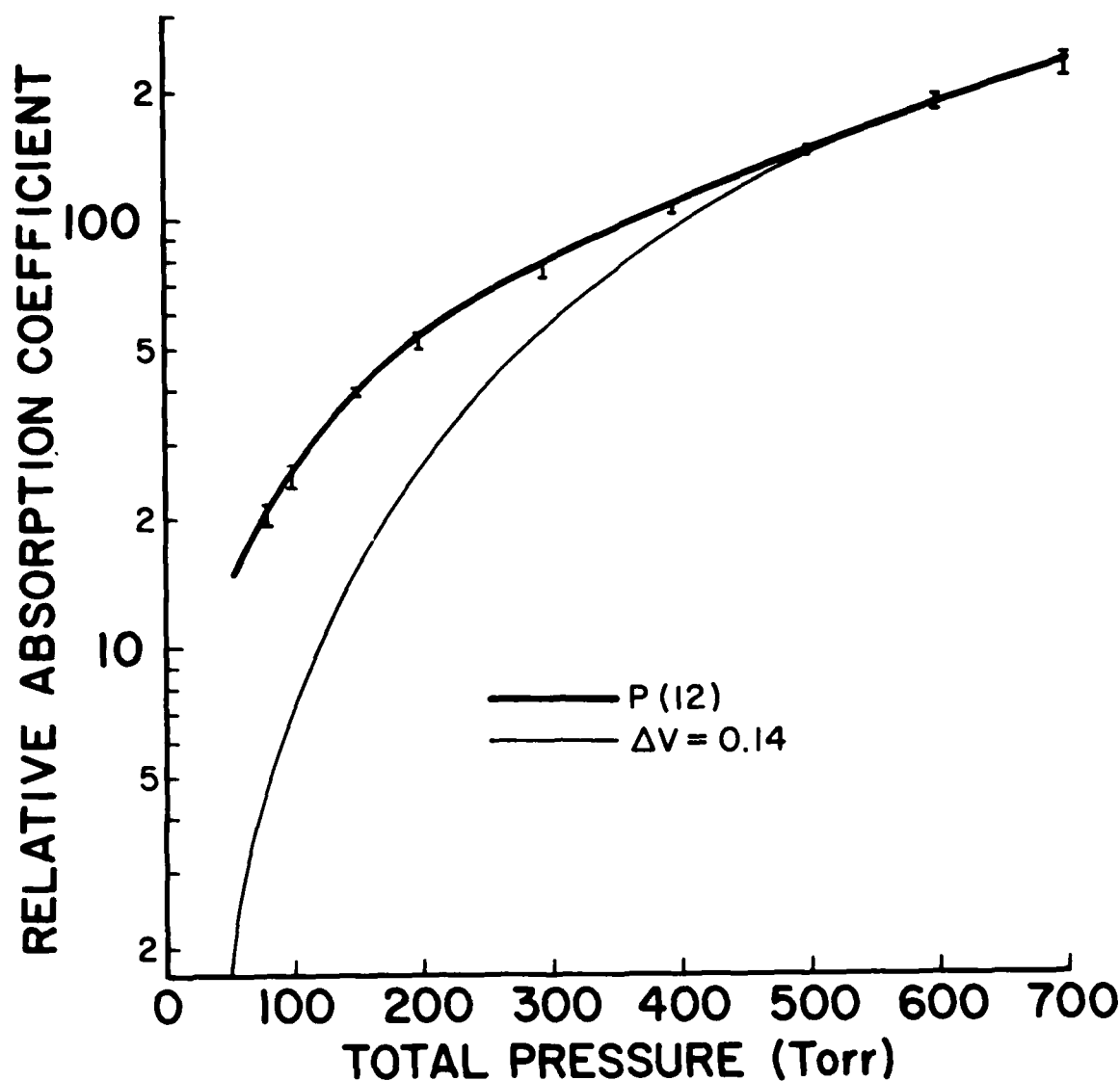


Figure 8. Experimental versus theoretical pressure dependence at the 10P(12) laser line for C_2H_2 absorption. The methodology employed was similar to that described in figure 7 except that only a single frequency model was used and no good agreement could be achieved. This indicates a much more complex structure than the $10\mu m$ P(10) pressure dependence curve.

The methodology described above offers possibilities for further refining experimental measurements of molecular absorption line center frequencies for trace gases in the atmosphere. Whereas spectrographic techniques (typically using a 3 to 5 m grating instrument) can produce frequency estimates to about 0.02 cm^{-1} ,* apparently pressure profiles can provide accuracies of better than about 0.001 cm^{-1} , a 20x improvement in resolution for absorption lines having a laser line center within 0.01 cm^{-1} . Even though a limited number of lines in a trace gas absorption spectrum may be close enough to laser lines to provide frequencies with accuracies improved by this technique, derived frequencies for a molecular series need verification most for a given member. Relative frequencies for transitions of a series usually involve less uncertainty.

3. CONCLUSIONS

We have presented in this report significant measurements of C_2H_4 absorption patterns of $9\mu\text{m}$ and $10\mu\text{m}$ C^{12}O_2 ¹⁶ laser transitions using the optoacoustic technology developed at the US Army Atmospheric Sciences Laboratory, White Sands Missile Range, New Mexico. This report also provides new data on the pressure dependency of ethylene as a trace gaseous absorber and demonstrates a technique for using this relationship to perform absorption spectroscopy with improved resolution.

This report discusses the continued discrepancy between experimental results in the measurement of C_2H_4 absorption coefficients for C^{12}O_2 ¹⁶ laser lines in the $9\mu\text{m}$ and $10\mu\text{m}$ bands. Discrepancies between our data and the calibration data of Patty et al⁸ of up to 30 percent (and from 10 to 27 percent between various other authors) raise serious concerns.

A number of measurements made with spectrophone systems (as used in this research) have been reported. Even on these measurements agreement is not consistent. Concrete explanations of the differences are rarely available, but the experience of this group indicates that purity of the constituents is of vital importance. Buffer gases and gas handling systems are likely sources of contamination. For the preceding measurements, buffer gas absorption was measured separately to insure functional purity; the gas handling system was of a very high quality (largely of stainless steel) and was effectively tested with the buffer gas check procedure.

*Estimate by ASL staff

⁸R. R. Patty et al, 1974, "CO₂ Laser Absorption Coefficients for Determining Ambient Levels of O₃, NH₃, and C₂H₄," Applied Optics, 13:2850

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