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Temperature dependence of line parameters of $^{12}C^{16}O_2$ near 2.004 μm studied by tunable diode laser spectroscopy*

Hongliang Ma(马宏亮)[1,2], Mingguo Sun(孙明国)[1], Shenlong Zha(查申龙)[2], Qiang Liu(刘强)[1], Zhensong Cao(曹振松)[1,†], Yinbo Huang(黄印博)[1], Zhu Zhu(朱柱)[2], and Ruizhong Rao(饶瑞中)[1]

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The absorption spectrum of carbon dioxide at 2.004 μm has been recorded at sample temperatures between 218.0 K and room temperature, by using a high-resolution tunable diode laser absorption spectrometer (TDLAS) combined with a temperature controlled cryogenically cooled absorption cell. The self-, N_2-, and air-broadening coefficients for nine transitions of $^{12}C^{16}O_2$ belonging to the 20012←00001 band in the 4987 cm^{-1}–4998 cm^{-1} region have been measured at different temperatures. From these measurements, we have further determined the temperature dependence exponents of the pressure-broadening coefficients. To the best of our knowledge, the temperature dependence parameters of the collisional broadening coefficients are reported experimentally for the first time for these nine transitions. The measured halfwidth coefficients and the air temperature dependence exponents of these transitions are compared with the available values reported in the literature and HITRAN 2012 database. Agreements and discrepancies are also discussed.

Keywords: carbon dioxide, absorption spectroscopy, temperature-dependent exponent, collisional broadening coefficient

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1. Introduction

Carbon dioxide (CO_2) is the major anthropogenic greenhouse gas that dramatically increased since the beginning of the industrialization era due to the effects of human activities on the Earth's environment.[1] Its infrared absorption and radiation play a crucial role in radiative transfer processes especially for the evolution of the Earth's atmosphere and climate. Therefore, accurate spectroscopic data, in particular in the near-infrared region, is essential, e.g., for reliable quantitative studies of global warming or analysis of remote sensing data.[2]

Currently, the global monitoring of CO_2 is of high importance and several methods[3–7] are developed for the monitoring of atmospheric carbon dioxide. Particularly, in 2009, the Greenhouse Gases Observing Satellite (GOSAT)[8] was launched by Japan to provide global time-dependent maps of CO_2 concentration in the 1.6-μm and 2.0-μm region. Therefore, the determination of the exact values of the spectroscopic parameters of the spectral lines around 2.0 μm is very important for the high precision measurements of CO_2 concentration in atmosphere, especially the temperature dependence exponents because of the wide temperature range in the Earth's atmosphere. Due to the importance in remote sensing applications, the absorption spectrum of CO_2 around 2.0 μm at different temperatures has been quantitatively studied by many groups.[9–14] In 1979, Valero *et al.*[9] reported the line intensities and half-widths of 51 lines at different temperatures for the 20013←00001 band of CO_2 at 4854 cm^{-1} with a Fourier transform spectrometer (FTS) with resolution of 0.075 cm^{-1}. Subsequently, in 1990, Suárez *et al.*[10] published the temperature dependence of self-broadened half-widths of CO_2 with an FTS and at a resolution of 0.004 cm^{-1} in the range 4850 cm^{-1}–5050 cm^{-1}. Recently, Joly L *et al.*[11] and Li J S *et al.*[12] determined the line strength, air-broadening half-width, air pressure-induced shift coefficient as well as their temperature dependences in the range 4870 cm^{-1}–4880 cm^{-1} of CO_2, in order to provide reference data for the differential absorption lidar instrument. Li J S *et al.*[13] also reported the self-induced pressure shift coefficients and the temperature dependence for 8 transitions in this region by a high resolution tunable diode laser absorption spectrometer combined with a cryogenically cooled optical multi-pass cell in 2012. More recently, the CO_2 line parameters including temperature dependences of air- and self-broadened line shapes near 2.06 μm have been studied by Benner *et al.*[14] Besides these results, no further low temperature experimental information has been obtained for these lines. However, the intercomparison data in Ref. [15] (Table 1, Table 3, and Table 4) show more measurements are needed for the CO_2–N_2 and CO_2–CO_2 systems, especially the temperature dependence exponents of CO_2 tran-

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sitions near 2.0 μm belonging to the 20012←00001 band, and hence the present measurements are important.

In the present work, we have investigated nine lines of CO_2 in the 4987 cm^{-1}–4998 cm^{-1} spectral region belonging to the 20012←00001 band, using a narrow linewidth tunable diode laser combined with a temperature controlled cryogenically cooled absorption cell. The temperature dependence exponent n and the pressure-broadened half-width coefficients for nine transitions of CO_2 with intensities stronger than 7.9×10^{-22} $cm^{-1}/(molecule \cdot cm^{-2})$ have been determined and compared with the available values reported in the literature and the HITRAN2012[16] (hereafter shortened to HITRAN) database.

2. Experiment details

The CO_2 molecular spectra in the 2.004-μm region are recorded with a high resolution tunable diode laser absorption spectrometer. The experimental setup is schematically shown in Fig. 1. A commercial distributed feedback (DFB) diode laser purchased from Nanoplus has been used as the light source. The side mode suppression ratio is higher than 40 dB. The diode laser emits radiation near 2.004 μm with no mode-hops over the tunable range. The typical output power is about 7.8 mW, with a relatively narrow linewidth of about 3 MHz, which can be neglected comparing to Doppler limited molecular absorption linewidth (for the CO_2 lines around 2.004 μm at 296 K, the typical Doppler half-width at half-maximum is about 230 MHz). The continuous tuning range (at constant temperature) is within 5 cm^{-1}. This point is of particular interest to reconstruct properly the baseline, as the laser sweeps the molecular transition over a spectral range that is large enough to yield zero-absorption signals at the end and at the beginning of the scanning interval. The laser wavelength is temperature-stabilized by means of a Peltier thermo-element and is driven by a low noise current supply (ILX Lightwave LDC-3724C). The controller was linked to a computer via a GPIB interface while its current was scanned in a step-by-step mode with typical size of 0.1 mA in order to scan the laser over the selected absorption lines by modulation of the driving current. The laser beam is usually collected by a fixed focus collimator and is separated into three parts via a wedged CaF_2 beam splitter. The main beam was passed through a sample cell for the absorption signal. The second beam was introduced onto a wavemeter (Bristol-621A) to record frequency of the absorption signal with the accuracy of 0.001 cm^{-1} at 2.0 μm, and the third one was used as a background signal. Two InGaAs amplified detectors (Thorlabs, PDA10DT-EC) were used to record the sample and the background spectra. A three-channel acquisition system ensures simultaneous recording of the gas sample absorption spectrum and the other two signals (wavemeter and background signal).

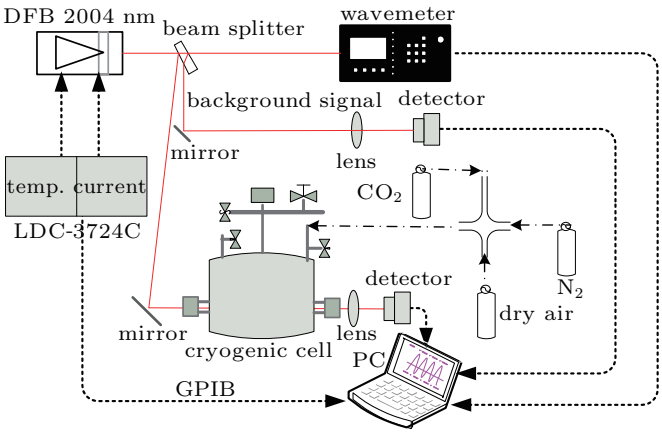


Fig. 1. (color online) The schematic diagram of the experimental setup for the CO_2 spectra measurement.

A home-made cooling cell was developed for the purpose of atmospheric applications, and more precisely described in recent publications.[17,18] Briefly, the absorption path length is 17.4 cm and the cooling system is realized by a dry nitrogen flow, circulating around the absorption cell. Two PT100 sensors mounted at different locations on the wall of the sample cell were used to measure the gas temperature during the recordings. The gas pressure is continuously measured by two capacitance gauges (Shanghai Zhentai CPCA-200, full range 20 kPa; MKS 626C, full range 67 kPa), and the pressures are measured with an uncertainty of 0.25%. The gas samples (carbon dioxide, air, nitrogen) were bought from Nanjing Special Gas Co., with a nominal purity of 99.99%.

Table 1. Summary of experimental conditions of CO_2 spectra analyzed in this work.

Temperature/K	CO_2 (VMR)	Pressure range/kPa
Pure CO_2		
296.1±0.3	1	11.446–28.064
258.2±0.2	1	12.827–28.118
253.4±0.1	1	12.120–30.837
243.7±0.3	1	9.526–31.424
232.7±0.3	1	10.159–27.144
218.4±0.3	1	10.456–25.425
CO_2 in air		
295.9±0.3	0.187	11.312–64.648
253.2±0.2	0.123	14.305–45.703
243.8±0.1	0.166	11.591–45.396
233.0±0.3	0.136	13.999–42.130
218.4±0.3	0.139	15.465–44.036
CO_2 in N_2		
296.2±0.3	0.176	11.456–64.808
253.2±0.2	0.154	12.200–38.770
243.8±0.1	0.161	12.546–47.276
232.9±0.2	0.135	12.464–37.277
218.4±0.3	0.160	15.612–44.343

During the measurements, the sampling cell was first evacuated to the order of 10^{-2} Pa using a mechanical together with a molecular pump (Leybold, TW300), then filled with the natural abundance CO_2 gas of 99.99% purity or the mixed gas

(dry air+CO_2 or N_2+CO_2). In order to mix the gases fully, absorption signals were always recorded after at least 3 hours of preparing the gas mixtures. All measurements were achieved with a temperature varying less than 0.3 K during a spectrum acquisition. For a given couple of pressure and temperature, 3–5 spectra were recorded to reduce systematic errors. Table 1 summarizes the experimental conditions for the entire set of spectra used in this work.

3. Data processing

For a monochromatic laser beam, the transmission of light radiation T_ν at a frequency ν through a uniform gas can be given by the Lambert–Beer law:

$$T_\nu = \frac{I_\nu(\nu)}{I_0} = \mathrm{e}^{-\kappa(\nu)L}, \tag{1}$$

where I_0 and $I_\nu(\nu)$ are referred to the incident and transmitted power, respectively, $\kappa(\nu)$ (in unit cm^{-1}) is the spectral absorption coefficient; L (in unit cm) is the optical path length within the cell.

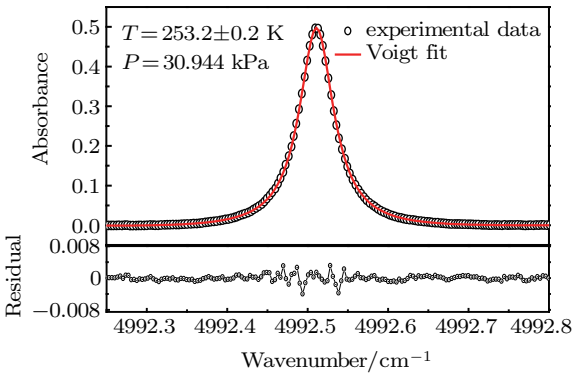


Fig. 2. (color online) Example of recorded absorption spectrum for the R(20) transition of CO_2 perturbed by air. Experimental profile and fitted Voigt profile are presented here. The observed minus calculated residuals are shown at the bottom. Pressure is 30.944 kPa and temperature is 253.2±0.2 K.

To retrieve the Lorentz half-width coefficients of these transitions of CO_2, the local baseline of the observed spectra was assumed to be a third order polynomial of the wavenumber and spectral lines were fitted with a Voigt function having four parameters, (line position, integrated absorbance, FWHM of the Gaussian and Lorentz components) that were determined using a multi-peak fitting program (http://www.unipress.waw.pl/fityk/). This software is an interactive nonlinear least-squares fitting program based on the Levenberg–Marquardt algorithm that could be simultaneously fitting a few lines for one spectrum at a time. During the fitting procedure, the FWHM of the Gaussian component of the lines were fixed to their theoretical values. The integrated absorbance were also fixed to the values that were calculated from line intensities (note that these line intensities were taken from HITRAN). Note that other weaker lines, belonging to the 21112←01101 band of CO_2, which need to be included in the nonlinear least-squares multi-peak fitting procedure for a correct determination of the line parameters. The four Voigt function parameters of these weaker lines were calculated from HITRAN. Figure 2 shows an example of the recorded spectrum and its corresponding Voigt fit.

The self-broadened half-widths were fitted assuming a linear pressure dependence using the relation:

$$\Delta\nu_\mathrm{L} = P_\mathrm{self} \cdot \gamma^0_\mathrm{self}. \tag{2}$$

In Eq. (2) γ^0_self is the retrieved broadening coefficient at 1 atm (1 atm = 1.01325×10^5 Pa), $\Delta\nu_\mathrm{L}$ is the measured broadened half-width of the spectral line and P is the total sample pressure.

For a given perturber, the collision half-width can be defined by:

$$\Delta\nu_\mathrm{L} = P_\mathrm{self} \cdot \gamma^0_\mathrm{self} + P_\mathrm{out} \cdot \gamma^0_\mathrm{out}, \tag{3}$$

where, P_self is the partial pressure of CO_2, γ^0_self is the coefficient for self-broadening, P_out is the perturber (air or nitrogen) pressure, γ^0_out is the coefficient for collisional broadening by air and nitrogen.

Using Eqs. (2)–(3), the broadening coefficients γ^0_self, $\gamma^0_{N_2}$, and γ^0_air are determined by plotting the Lorentz widths versus gas pressure and then fitting them by straight lines. The collisional broadening coefficient at a given temperature is expressed as the slope of the best fit line. Figure 3 illustrates the plot of the measured air-broadened half-widths versus pressure at 218.4 K for the R(20) line in the 20012←00001 band of CO_2. The same procedure is also followed for the other transitions.

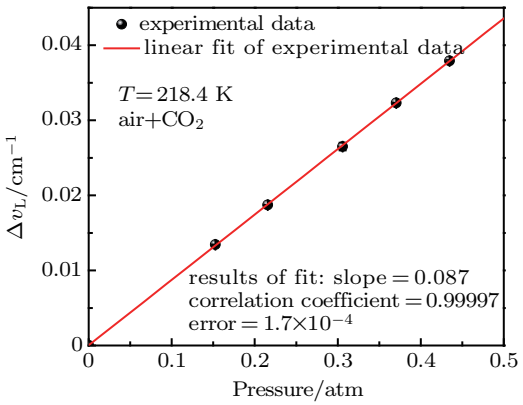


Fig. 3. (color online) Plot of collisional half-width at half maximum versus each pressure for the R(20) transition of CO_2. The slope of the regression line corresponds to the air-broadening coefficient in $\mathrm{cm}^{-1}\cdot\mathrm{atm}^{-1}$ at 218.4 K.

The temperature dependence of the half-width is expressed using the power law model, and is generally expressed as:

$$\gamma(T) = \gamma(296\ \mathrm{K}) \left[\frac{296\ \mathrm{K}}{T}\right]^n, \tag{4}$$

where $\gamma(296\ \mathrm{K})$ is the broadening coefficient of the line at 1-atm pressure and reference temperature T = 296 K, and

$\gamma(T)$ is the retrieved broadening coefficient at 1atm pressure and temperature T; n is the temperature dependence exponent. Figure 4 shows the air-broadening coefficient temperature dependence exponent of the R(28) transition of CO_2 as an example.

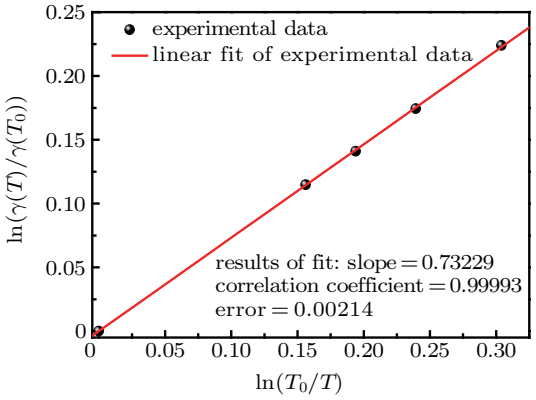


Fig. 4. (color online) Fits $\ln[\gamma(T)/\gamma(T_0)]$ of as a function of $\ln(T_0/T)$ for the R(28) line of CO_2 using air-broadened widths measured over the range 218.4 K–295.9 K.

4. Results and discussion

Within the tunable range of the Nanoplus diode laser, nine strong lines of CO_2 in the 20012←00001 band lying between 4987 cm^{-1} and 4998 cm^{-1} were investigated in the laboratory.

4.1. Self-, N_2-, and air-broadening coefficients

In Fig. 5 the broadening parameters obtained with the Voigt profile are plotted as a function of the rotational quantum number m ($m = J + 1$ for R branch lines) at various temperatures for the R(12) to R(28) of 20012←00001 band of CO_2. In Fig. 5, we have noticed that: firstly, all the pressure-broadening coefficients increase with the diminution of the temperature; secondly, $\gamma^0_{self} > \gamma^0_{N_2} > \gamma^0_{air}$ at the same temperature was noticed from the dataset; third, all the results present similar m-dependences. The error bars are one standard deviation given by the linear regression. However, these values are not considered to represent the absolute uncertainties because there may be systematic uncertainties arising from additional sources, such as those related to the knowledge of wavenumber calibration errors, gas sample pressures and normalization uncertainties. The absolute accuracies in our measured parameters due to various errors are estimated to be better than 4%, as stated from our experience.

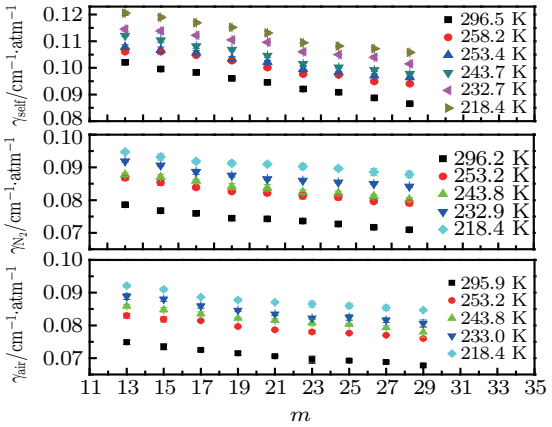


Fig. 5. (color online) Self-, N_2-, and air-broadening coefficients as a function of the rotational quantum number m at various temperatures for R(12) to R(28) of 20012←00001 band of CO_2.

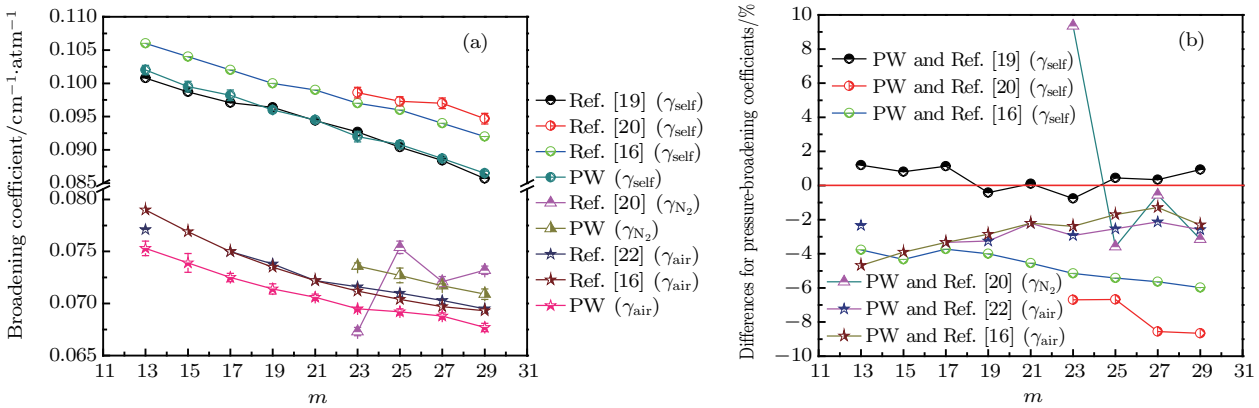


Fig. 6. (color online) (a) Comparison between our values of pressure-broadening coefficients in the 20012←00001 band of CO_2 obtained at room temperature and previous results obtained by other authors[19,20,22] and HITRAN.[16] (b) The percent differences {(Present work–other results)/other results ×100%} of pressure-broadening coefficients between this work and the results from Refs. [19], [20], [22], and HITRAN[16] for transitions in 20012←00001 band are plotted as a function of the m value (Note: PW = present work).

In order to assess the quality of our retrieved results, we compared our broadening coefficients at room temperature with other corresponding measurements[16,19–22] for this band of CO_2 transitions, shown in Fig. 6(a). To extend our comparison in Fig. 6(b) we have plotted against m the percent differences {(Present work–other results)/other results×100%} between our values and other reported results. Firstly we compare our self-broadening coefficients with previous determinations of Régalia-Jarlot *et al.*,[19] Corsia *et al.*,[20] and HITRAN.[16] As shown in Fig. 6, our values are close to those of Régalia-Jarlot *et al.*,[19] the difference lies within −0.76% to 1.19% and a value of 7.3×10^{-4} cm^{-1}·atm^{-1} is achieved for the RMSD (root-mean-square error) of our results and the measurements reported by Régalia-Jarlot *et al.*[19] We can see

that the difference between our self-broadening coefficients and the values from the HITRAN database varies from −3.7% to −6.0% for all the lines (for these transitions, the HITRAN database has assumed the self-broadening coefficients from Ref. [21]). However, the differences between our results and HITRAN (or Ref. [21]) maybe attributed to the different setup and the analysis techniques employed in the two studies. It can be noticed that in Ref. [21], the self-broadening coefficients were obtained using an FTS and the spectral resolutions for their spectra are 0.010 cm^{-1} to 0.013 cm^{-1} while we were using a very narrow laser source with a resolution of about 0.0001 cm^{-1}. When comparing our results (for R(22), R(24), R(26), R(28)) with those from Corsia *et al.*[20] we found that the difference becomes large (up to −8.66%), namely, our results are systematically lower than those obtained by Corsia *et al.*[20] However, we note that the data from Corsia *et al.*[20] are clearly higher for the transitions (R(22), R(24), R(26), R(28)) in comparison with all results. At present, we cannot explain these different findings, maybe due to unknown reasons.

As to N_2-broadening coefficients, we could find only the values listed by Corsia *et al.*[20] for direct comparison. Only four lines are identical to the two works. For the transitions R(24), R(26), and R(28), the differences are −3.58%, −0.55%, and −3.14%, respectively. Regarding the R(22) line, the discrepancy in N_2-broadening coefficient between our measurement and the result from Corsia *et al.*[20] is up to 9.36%. It is difficult to explain this rather large difference for the transition. However, it is interesting to note that, the self-broadening coefficient and O_2-broadening coefficient of the R(22) line are always larger than the other three lines (R(24), R(26), and R(28)) as shown in Ref. [20].

For air-broadening coefficients, we have also compared our values with those from HITRAN and Toth *et al.*[22] Our results of measured air-broadening coefficients are much closer to those of HITRAN and the results of Toth *et al.*[22] The agreement between our values and those obtained by Toth *et al.*[22] is better than 3.4% for all transitions. (Note: the air-broadening coefficient of R(14) was not given in Ref. [22].) With the exception of R(12), our broadening parameters agree within 4.0% with those from HITRAN. The RMSD of our results and the measurements reported by Toth *et al.*[22] and HITRAN are 2.0×10^{-3} $cm^{-1}\cdot atm^{-1}$ and 2.2×10^{-3} $cm^{-1}\cdot atm^{-1}$, respectively.

4.2. Temperature dependence exponent

For remote sensing applications, both temperature and pressure dependences of half-width coefficients are required on a line-by-line basis. In determining the temperature dependence exponents for the Lorentz self-, air-, and N_2-broadened half-width coefficients, we have employed the power law relationship given in Eq. (4) by fitting the pressure-broadening coefficients at all sample temperatures. The results are summarized in Table 2. The uncertainties in parentheses represent only the uncertainties as obtained from the linear regression. The absolute uncertainties of the temperature dependence exponents are difficult to assess, but are less than 10%, as considering all the unknown sources of errors. From Table 2, unlike broadening coefficients, we can see that the temperature dependence exponents are less dependent on m values. The mean values of the temperature dependence exponent for self-, air-, and N_2-broadening coefficients are 0.685, 0.660, and 0.714, respectively. Meanwhile, we also compared our air temperature dependence exponents with the values from HITRAN, as it is shown in Table 2. The RMSD of our air temperature dependence exponents and the results given in HITRAN is 0.0153, while the agreement is better than 2% for most of the transitions. Although the range in $n_{\mathrm{EXP}}^{\mathrm{air}}$ and n_{H} is generally close, there is less scatter in the results of HITRAN. Yet, the dependence in the rotational quantum number m of HITRAN is a theoretical calculation, while our experimental results were affected by other factors. Unfortunately, we did not find the self (or N_2) temperature dependence exponents for comparison.

Table 2. Self-, air-, and N_2-temperature dependence exponents for the R(12) to R(28) of 20012←00001 band of CO_2. The air temperature dependence exponent from HITRAN also presented in the table and compared with those of our results. Numbers in parentheses of our work are the error obtained on the linear regression.

Line	m	ν_0/cm^{-1}	n				$(\frac{n_{\mathrm{exp}}^{\mathrm{air}}-n_{\mathrm{H}}}{n_{\mathrm{H}}})\%$
			This work			HITRAN	
			self	N_2	air	air	
R(12)	13	4987.30835	0.613(20)	0.620(13)	0.695(9)	0.69	0.72%
R(14)	15	4988.65479	0.580(16)	0.662(12)	0.722(12)	0.69	4.64%
R(16)	17	4989.97152	0.699(6)	0.637(5)	0.697(16)	0.69	1.01%
R(18)	19	4991.25851	0.742(7)	0.666(8)	0.695(9)	0.70	–0.71%
R(20)	21	4992.51574	0.698(13)	0.649(10)	0.703(10)	0.71	–0.99%
R(22)	23	4993.74317	0.714(11)	0.646(15)	0.729(13)	0.72	1.25%
R(24)	25	4994.94078	0.710(13)	0.676(11)	0.732(10)	0.73	0.27%
R(26)	27	4996.10854	0.717(13)	0.688(12)	0.717(4)	0.74	–3.11%
R(28)	29	4997.24641	0.694(10)	0.696(14)	0.732(2)	0.75	–2.40%

Note: The wave number values ($\nu_0(cm^{-1})$) are extracted from HITRAN; $n_{\mathrm{exp}}^{\mathrm{air}}$ are the air temperature dependence exponents from this work; n_{H} are the air temperature dependence exponents from HITRAN; $(\frac{n_{\mathrm{exp}}^{\mathrm{air}}-n_{\mathrm{H}}}{n_{\mathrm{H}}})\%$ are the percent differences between our results and HITRAN.

5. Summary

We measured nine CO_2 self-, N_2-, and air-broadening half-width coefficients for the 20012←00001 band in the 4987 cm^{-1}–4998 cm^{-1} region using a narrow linewidth diode laser associated with a home-made cryogenic cell. The results show that the broadening coefficients present clear temperature- and *m*-dependences. Furthermore, the parameters obtained at room temperature were thoroughly compared to former studies and the HITRAN database, and some agreements and discrepancies have been found. Self-broadening coefficients were determined for nine transitions and found to be from 3.7% to 6.0% lower than those from HITRAN, but very close to those obtained by Régalia-Jarlot *et al.*[19] For N_2-broadening coefficients, only 4 lines are available in Ref. [20] for a direct comparison. The results show that good agreement was achieve between our values and those from Corsia *et al.*[20] except R(22). We have also compared our air-broadening coefficients with those from HITRAN and Toth *et al.*[22] The agreement is within 3.4% (nine lines for Toth *et al.*[22]) and 3.5% (except R(12) and R(14) lines for HITRAN) for most of the transitions, respectively. From these broadening coefficients at different temperatures, the temperature dependence exponents have been determined and no significant tendency was found. Our air temperature dependence exponents agree very well with those in HITRAN with an RMSD of only 0.0153. To the best of our knowledge, the temperature dependence exponents of the self-, N_2-, and air-broadening coefficients are reported experimentally for the first time for these nine transitions of CO_2. We believe that the reported data set presented here will be helpful for the Earth's radiation balance or remote sensing of CO_2 in the atmospheric spectral window at 2.0 μm.

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