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Reinvestigation of pressure broadening parameters at 60-GHz band and single 118.75 GHz oxygen lines at room temperature

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Abstract

Low pressure measurements of broadening parameters of the 118.75 GHz fine structure line of oxygen molecule have been made by a BWO-based spectrometer with acoustic detector (RAD) at room temperature. Pressure broadening parameters were obtained for the buffer gases O_2 , N_2 ,

Keywords: Oxygen; Fine structure; Line broadening; Line center; Microwave spectroscopy; Atmosphere propagation model

1. Introduction

Oxygen millimeter wave spectra belong to a series of magnetic dipole transitions within $^3\Sigma$ ground electronic state. Each rotational level is split by spin–spin and spin–orbit interactions which makes for total angular momentum N take the values $N^\pm=J,J\pm1$. Within each triplet labeled by J there are two allowed $(N,J)-(N,J+1)=N^+$ and $(N,J)-(N,J-1)=N^-$ fine structure transitions. These lines mostly fall at 60-GHz frequency range and one line 1^- at 118.75 GHz is isolated.

They are used for Earth atmosphere remote sensing purposes and for astronomical observation of molecular oxygen, e.g. in airborne NAST-M passive imaging microwave spectrometer [1], considered for use in Geostationary Observatory for Microwave Atmospheric Sounding (GOMAS) [2]. The EOS MLS apparatus focus on temperature and pressure sensing using the line at 118-GHz [3]. And 118.75-GHz fix-tuned HEMT preamplifier has been installed in recently launched satellite

*Corresponding author. Fax: +7-8312-36-3792. E-mail address: glb@appl.sci-nnov.ru (G.Yu. Golubiatnikov). ODIN to allow very sensitive searches for interstellar O₂ [4] and used for retrievals of the atmospheric temperature vertical profile [5].

As it has been shown in our recent re-measurements [6] of rotational line (N,J)=(3,2)-(1,2) at 425 GHz by the RAD spectrometer the broadening coefficients of this line significantly (about 25%) differ with previously measured ones for oxygen and nitrogen perturbers at room temperature in [11]. So we have continued our measurements of line parameters for another oxygen transition. In the paper [7], we have reported the oxygen central frequency measurements and broadening coefficients in O_2 and N_2 for eight oxygen rotational lines up to 1.12-THz, which also showed better accuracy and internal consistency than previous measurements.

In the present paper, we report the results of laboratory measurements by the RAD spectrometer of the central frequencies and collisional broadenings of some fine structure oxygen transitions at room temperature. The most extensive studies were done on the line at 118.75 GHz, where broadenings by O₂, N₂, H₂O, CO₂, CO, Ne, He, and Ar as buffer gases were investigated. The motivation in the choice of gases was defined by the

atmospheric applications of the line studied, and the intention to obtain experimental data for possible theoretical interpretation of the collisional relaxation of magnetic-dipole transitions. Broadening parameters by O_2 , N_2 and line center measurements of lines 1^+ , 5^- , 7^+ , 11^+ , and 15^- at 60-GHz frequency band were also measured. These measurements together with the previous of ours and another authors studies present more or less complete material for comparison of the broadening parameters of different types of oxygen molecular lines (fine structure, rotational, and electronic) ranging from millimeter to optical wave lengths and their dependencies on quantum numbers (J, N).

Well-known difficulty in the studies of oxygen lines is weakness of transitions induced by magnetic dipole moment; another one is extreme sensitivity of the transitions to the Earth magnetic field. To overcome these difficulties many different techniques were used.

Self-broadening of the line at 118.75 GHz was studied earlier in [8–18] by different methods as by low-pressure microwave spectroscopy with magnetic modulation and by the conventional absorption measurements in a long (500 ft) cell up to atmospheric pressure. Much less studies were done to investigate foreign broadening of this line [9,11,14,18] evidently due to the weakness of the line. Broadening parameters of the 60-GHz band lines were studied earlier most thoroughly in [29,30] by inter-resonator (two-resonator dispersion detection) spectrometer at pressures <20 Torr. Some measurements were done in [13,27] by low-pressure microwave spectroscopy with magnetic modulation and in [25] by resonator spectrometer by changing of magnitude of frequency deviation. Resulting lineshape of the 60-GHz group and line mixing coefficients were given most completely in [8]. Some of the measurements were in disagreement, e.g., the self-broadening for fine structure line 5⁻ at 60.306 GHz was measured in [25] as 2.4 MHz/Torr against 1.89 MHz/Torr in [29], which amounted in more than 20% difference.

Measurements by the RAD technique (direct absorption in a small acoustical cell) reported in the present paper give possibility to compare results of different techniques and to have more reliable error limits. Advantages of the RAD spectrometer were recording of the true absorption profiles of the lines by means of amplitude modulation with signal-to-noise ratio (SNR) better than 100, which was demonstrated earlier in studying rotational spectrum of oxygen molecule [6,7]. Improving of accuracy of the broadening parameters is important for more accurate modeling of experimental lineshapes for retrieval of atmospheric temperature profiles [5].

2. Experimental details

A BWO-based spectrometer was used with acoustic detection (RAD) [19]. The study of the rotational

spectrum of oxygen lines in submillimeter wave range [6,7] has demonstrated that the accuracy for such measurements can be significantly improved by using RAD. The double magnetic shielding of the gas cell was used, because oxygen magnetic-dipole transitions are sensitive to the Earth's magnetic field. The shielding case was made of annealed permalloy. The high power of BWO (output power was about 40 mW), phase locked against reference synthesizer, also permitted to increase sensitivity of the spectrometer. Typically, lines were recorded by amplitude modulation technique with SNR of the order 200. For strong lines of 60-GHz group SNR was more than 350 and even for some weak lines in N₂ it was no less than 50. The record of one line took about 2–4 min (integration time was 1–2 s/frequency point).

The block-scheme and details of such spectrometer were given in [6,7]. In the present study, BWO sources as OB-86 in permanent magnet of the frequency range 110–180 GHz and OB-70 of the range 52–79 GHz were employed. Whilst in the present study, it was used completely different frequency range in comparison with [6,7], where lines up to 1.12 THz were studied, nevertheless the spectrometer arrangement—synthesizer, diode mixer, PLL system, and acoustic cell as detector were all the same. That is a great advantage of RAD spectrometer, which covers the whole millimeter and submillimeter bands by just the change of the radiation source (in our case BWO with horn and some waveguides).

The partial pressure of O_2 , first filled into the sample cell, was about 1 Torr to get sufficient SNR, and the selfbroadened halfwidth of the line was determined. Then, a controlled amount of foreign gas was added and the line halfwidth was determined again. Further adding a foreign gas up to partial pressures of 3-4 Torr continued this procedure. To stabilize the pressure in the gas cell, we used additional buffer volume in vacuum system (a quartz chamber of about 10 times larger in volume than the gas cell). That was important for measurements of broadenings by water vapor, which adsorbed by cell walls quite much. The measurements were started only when the pressure in the cell was stable (usually we waited 15–20 min). For checking that the ratio between partial pressures of absorbing sample gas and water vapor corresponds to expected value, we changed the turn of gas filling (oxygen-water vapor, water vapor-oxygen). This check was also done for different ratios between partial oxygen and water vapor pressures. The obtained corresponding line halfwidths occurred to have the same values.

The gas pressure in the cell was measured by MKS Baratron gauge. The gases were from Messer MG Company with 4.5–5.0 grades of purity. Water sample was obtained from double distilled water source. Temperature was measured near the gas cell with uncertainty \pm 1 °C during an experiment.

The spectrometer sensitivity is in direct proportion to source radiation power. This advantage of RAD spec-

trometer was used for detection of weak oxygen lines. But BWOs output power is frequency dependent, and also modified by interference between incident and reflected radiations (reflections occur in cell windows, waveguide parts, etc.) makes also effect on absorbed power. Such frequency dependence should be taken into account as multiplicative term to the lineshape function. Moreover, the high power of BWO used in the present study (about 40 mW), weakness of oxygen lines and the technique of acoustic detection gave noticeable parasitic signal due to absorption of radiation in dielectric windows of the cell. So additive terms as fringes in baseline of conventional absorption spectrometer should be also taken into account. Thus, the model function to be used for the fit of experimental line should have both multiplicative and additive terms to the lineshape profile:

$$I(v) = A_0 \cdot (1 + A_1 \cdot (v - v_0)) \cdot \Gamma(v - v_0) + A_2 + A_3 \cdot (v - v_0) + A_4 \cdot (v - v_0)^2,$$
(1)

where A_0 is amplitude factor proportional to incident radiation power; the coefficient A_1 corresponds to frequency dependence trend of the BWO output power and interference effects; ν is the frequency; ν_0 is the line center frequency; $\Gamma(\nu-\nu_0)$ is normalized line profile function, and A_2 , A_3 , and A_4 are first coefficients of polynomial series describing baseline fringes.

The following measurements were also made—base-line record with the cell filled by nitrogen at different cell positions and with varying BWOs power. Then, for the same conditions and the same pressures we measured oxygen line. Then, we subtracted baseline and made a fit and compared obtained linewidths with ones obtained by using model of Eq. (1). It was found that linear and quadratic with frequency terms $(A_3, A_4 \text{ in Eq. (1)})$ modeling the baseline were sufficient for correct linewidth retrieval.

It should be also noted that for fittings of 60-GHz and 118-GHz lines we used Lorentz profile instead of Voigt function (Doppler widths are 129 kHz for 118 GHz line and 66 kHz for 60-GHz band at room temperature). So far the difference in line halfwidth determination by using Voigt and Lorentz model function was 5-6 kHz (at 1 Torr pressure for 118-GHz line) which was within experimental error. For pressure broadening coefficient determination the difference would be even lesser as 2-3 kHz/Torr. As an example, the record of the 11⁺ oxygen line at 1.69 Torr pressure with the residuum of the fit to Lorentz model Eq. (1) is shown in Fig. 1.

3. Results and discussion

The oxygen collisional linewidth (HWHM) was determined from the fit of the experimental lineshape to the Lorentzian line profile as is shown in Fig. 1, where the intensive 11⁺ line belonging to 60-GHz band is de-

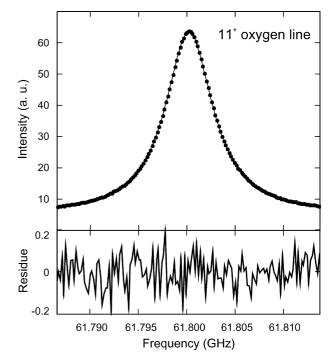


Fig. 1. Record of the 11^+ oxygen line. Experimental points are denoted by solid circles, the line represents the fit to Lorentz line profile; bottom trace is residuum of the fit. Line center is 61 800.157(7) MHz; pressure in the cell was $P=1.69\,\mathrm{Torr}$ at 298 K.

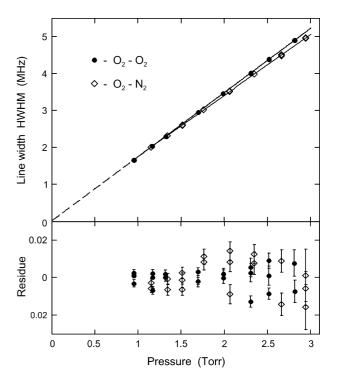


Fig. 2. Self- and N_2 -broadened widths (HWHM) vs. pressure of line 11^+ oxygen line at temperature 297 K. Experimental points are denoted by circles for O_2 , slope is $1.735(10)\,\text{MHz/Torr}$, intercept is $+25\,\text{kHz}$; by diamonds for $N_2-1.650(30)\,\text{MHz/Torr}$, $+115\,\text{kHz}$ (note about the intercept, see the text). The residua of the fits with error bars are shown below.

picted. Good SNR>350 reveals in residue validity of line function model Eq. (1) used in the present study. That also shows advantages of the acoustic detection technique to determine accurately lineshape parameters.

The widths (HWHM) of 11^+ line broadened by O_2 and N_2 plotted against total pressure, fittings to the pressure linear dependence function, and the residua of the fits are presented in Fig. 2. Residues show good accuracy and reproducibility. Systematic and statistical errors were of about the same order of magnitude. Intercept point of regression line with zero pressure axis for self-broadened line is $+25 \, \text{kHz}$, which means the low residual Zeeman and other line broadening effects.

Examples of the 1⁻ line at 118.75 GHz broadened by O_2 , and foreign perturbers as Ne and H_2O are presented in Fig. 3. Intercept for self broadened line is $+25 \, \text{kHz}$. Linewidth (Δv) of gas mixture is determined as $\Delta v = \gamma^{a-a} p_a + \gamma^{a-b} p_b$, where p_a and p_b are partial pressures of absorber and buffer gases, γ^{a-a} and γ^{a-b} are selfand foreign-broadening parameters. So intercept point (at p=0 Torr) in the case of gas mixture depends very

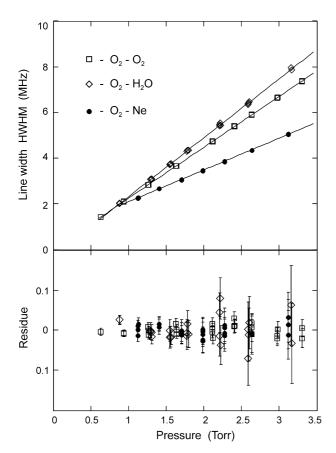


Fig. 3. The 118.75-GHz linewidths (HWHM) self-broadened (boxes), broadened by Ne (circles), and by H_2O (diamonds). Linear regression fits to the data are depicted by solid lines. Slopes are 2.230(10), 1.375(20), and 2.520(40) MHz/Torr, intercepts at p=0 Torr are +25, +740, and -170 kHz correspondingly (see text). The residua of the fits are shown below.

much on initial pressure of absorber gas and on the ratio of $\gamma^{a-a}/\gamma^{a-b}$ and have no physical meaning.

The broadening parameters of 118.75 GHz line obtained in this work and previously measured ones for 118.75 GHz line are listed in Table 1. In many cases values obtained by different authors agree within the errors quoted, which may give some confidence in reliability of the measurements. To the authors' knowledge, the linewidths broadened by CO, CO₂, and Ne for this line were measured for the first time. The linewidths in nitrogen exceed slightly those in oxygen except data from [14]. But as it is shown below in Tables 2 and 3 broadenings by nitrogen decrease faster in comparison with broadenings by oxygen with increasing quantum numbers.

Literature broadening parameters for microwaves and optical ranges are listed in Table 2. We try to sort out literature data to represent and compare different types of transitions, and not to overburden Table 2. Thus, it includes hyperfine structure lines [11,29,30], pure rotational transition [7,28], the infrared lines of $a^1\Delta_g$ – $X^3\Sigma_g^-$ (v=0–0) band [23], submillimeter transition

Table 1 Self- and foreign gas broadening parameters for 118.75-GHz oxygen line at room temperature; comparison with previous measurements

Pertuber	Broadening parameter (MHz/Torr)	Temperature (K)	References
O_2	2.230(10)	295	This work
	1.99(10)	300	[12]
	1.97	300	[13]
	1.94(30)	300	[14]
	1.98(07)	295	[10,11]
	2.19(11)	300	[15]
	2.13(04)	298	[9]
	2.11(05)	300	[16]
	2.14	300	[17] (calc.)
N_2	2.245(20)	297	This work
	1.75(25)	300	[14]
	2.20(10)	298	[9,11]
Ar	2.005(20)	296	This work
	2.01(09)	298	[9]
Ne	1.375(20)	296	This work
He	1.620(30)	297	This work
	1.61(06)	298	[9]
H_2O	2.520(40)	296	This work
	2.30(24)	298	[9]
CO_2	2.660(80)	296	This work
CO	2.310(50)	296	This work
Air	2.240(20)	293, dry air	This work (calc.)
	2.251(20)	293, 30% of H ₂ O	This work (calc.)
	2.14(07)	294, dry air	[18]
	2.17(07)	300, natural air	[8]
	ل 1 1 , س	500, natural an	r _c 1

Table 2 Microwaves and optical measurements of pressure broadening parameters of some oxygen lines taken from the literature

Transition	Broadening (MHz/Torr)	T (K)	Perturber	n	References
1-	1.98(7)	295	O_2	1.0(2)	[11]
1-	2.20(10)	298	N_2	0.7(2)	[11]
PP 1,1 A band	2.319(037)	296	O_2	0.67	[20]
PP 1,1 A band	2.339(035)	296	N_2	0.77	[20]
PP 1,1 B band	2.15(0.04)	296	O_2		[22]
PP 1,1 γ band	2.29(400)	296	O_2		[22]
$^{r}R(1), a^{1}\Delta_{g} - X^{3}\Sigma_{g}^{-}$	2.51(470)	296	O_2		[23]
5,4–3,4	2.010(060)	300	O_2		[7]
PP 5,5 A band	2.035(14)	296	O_2	0.70	[20]
PP 5,5 A band	2.075(025)	296	N_2	0.71	[20]
PP 5,5 B band	2.047(028)	296	O_2		[22]
PP 5,5 γ band	2.118(130)	296	O_2		[22]
7,6–5,6	1.94(060)	302	O_2		[7]
7-6, $a^{1}\Delta_{g}$	2.006(64)	290	O_2	-0.11	[24]
7–6, $a^{1}\Delta_{g}$	1.81(08)	290	N_2	0.6	[24]
$^{p}P(7), a^{1}\Delta_{g}-X^{3}\Sigma_{g}^{-}$	1.87(27))	296	O_2		[23]
$^{r}R(7)$, $a^{1}\Delta_{g}-X^{3}\Sigma_{g}^{s}$	2.07(07)	296	O_2		[23]
PP 7,7 A band	1.968(026)	296	O_2	0.71	[20]
PP 7,7 A band	2.008(006)	296	N_2	0.74	[20]
9+	1.79(2)	300	O_2	0.8	[30]
9,9-7,8	1.703(052)	293	O_2		[28]
9,9–7,8	1.32(23)	293	N_2		[28]
PP 9,9 A band	1.909(023)	296	O_2	0.71	[20]
PP 9,9 A band	1.934(015)	296	N_2	0.72	[20]
15,14–13,14	1.715(052)	293	O_2		[28]
$^{p}P(15), a^{1}\Delta_{g}-X^{3}\Sigma_{g}^{-}$	1.8(2)	296	O_2		[23]
$^{r}R(15)$, $a^{1}\Delta_{g}^{s}-X^{3}\Sigma_{g}^{\frac{s}{2}}$	2.00(7)	296	O_2		[23]
PP 15,15 A band	1.783(029)	296	O_2	0.66	[20]
PP 15,15 A band	1.775(021)	296	N_2	0.68	[20]
PP 15,15 B band	1.72(04)	296	O_2		[22]
PP 15, 15 γ band	1.70(14)	296	O_2		[22]

n is temperature dependence exponent of the broadening.

In [8], n = 0.8 is given for all fine structure transitions and used in atmospheric absorption model (MPM92).

Table 3
The fine structure linewidths broadened by O_2 and N_2 at room temperature, and comparison with previously measured data

N^{\pm}	Line center (MHz)	O ₂ -O ₂ broadenings (MHz/Torr)		This work (MHz/Torr)		
		Ref. [29]	Ref. [27]	Ref. [13]	$\overline{\mathrm{O}_2}$	N_2
1-	118750.340(7) ^a	2.11		1.97	2.230(10)	2.245(20)
1+	56264.785(40) ^a	2.22			2.21(04)	2.27(10)
3-	62486.26	2.00	1.96			
3+	58446.59	1.97	1.96	2.07		
5-	60306.052(8) ^a	1.89	1.60	2.4 [25]	1.900(20)	1.910(25)
5+	59590.98	1.86	1.56	1.80		
7-	59164.20	1.83	1.70	2.01		
7+	60434.777(8) ^a	1.79	1.68		1.810(15)	1.800(25)
9-	58323.88	1.76	1.64	1.94		
9^{+}	61150.56	1.74	1.42			
11-	57612.48	1.71				
11+	61800.157(7) ^a	1.70	1.60		1.735(10)	1.650(30)
13-	56968.21	1.67				
13+	62411.21	1.66	1.54			
15-	56363.391(7) ^a	1.63			1.695 (20)	1.580(40)
15+	62997.98	1.63			` '	. ,
17-	55783.80	1.60				
17+	63568.52	1.60	1.50			

Line center frequencies obtained in the present work are also given.

In [13] data were obtained at $T = 300 \,\mathrm{K}$; in [29] broadening of $\mathrm{O_2-O_2-at}$ 300 K (uncertainties were about 3%); in [25]— $T = 296 \,\mathrm{K}$; these work data obtained at $T = 297-298 \,\mathrm{K}$.

^a The line center frequencies have been measured in this work.

of $a^1\Delta_g$ state [24], and optical measurements of electronic states in A, B, and γ bands [20–22].

Temperature dependence exponent n is also listed in Table 2. Note that linewidth versus temperature is expressed by the empirical relation $\Delta v(T) = \Delta v(300 \text{ K}) \cdot (300/T)^n$, where n is temperature exponent.

In Table 3, the broadenings of 1+, 1-, 5-, 7+, 11+, and 15⁻ lines by O₂, N₂, measured by us, are presented together with previous measurements of other authors [13,25,27,29]. Measurements of line center frequencies of the 1^+ , 1^- , 5^- , 7^+ , 11^+ , and 15^- of ${}^{16}O^{16}O$ lines are also given. The listed frequencies are within experimental uncertainties of earlier measurements (see, for example [26,29,31]), but we would like to note the internal consistency of the present measurements with our predictions given in Table 4 of [7]. The 118750.340(7), 61 800.157(7), 60 434.777(8), and 56 363.391(7) MHz lines have discrepancy with our calculations in [7] no more than 2 kHz, 60 306.052(8) and 56 264.785(40) MHz lines have discrepancies with -4 and +9 kHz, respectively. We did not observe the pressure induced line shifts in the limits of $\pm 10-20 \,\mathrm{kHz/Torr}$.

Obtaining in the present and other recent works of the accurate broadening parameters of oxygen molecule lines ranging from millimeter (fine structure) through submillimeter (rotational) up to the optical (electronic) wave bands gives the possibility to compare both values of broadening parameters and their dependencies on quantum numbers. In Fig. 4, the self-broadening coefficients for oxygen lines are plotted as function of quantum number (N, J). Together with the present data, the values are taken from [29] (60-GHz), rotational broadenings from [7], and electronic states A band from

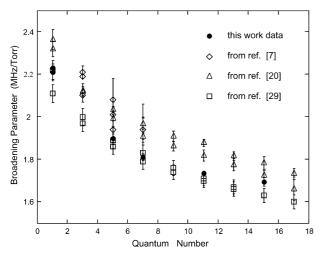


Fig. 4. Comparison of experimental self-broadening parameters at room temperature for different oxygen spectra: fine structure lines [29] (boxes) and present work measurements (solid circles), rotational [7] (diamonds), electronic PP and RR transitions [20] (triangles) as a function of quantum number $N^{\pm} = J \pm 1$. For pure rotational lines quantum numbers are used of upper state N, in [20] quantum numbers are also given for upper state N = J.

[20]. Inspection of Fig. 4 reveals close similarity of broadenings and their dependencies on rotational quantum numbers for these very different oxygen spectra. The average difference between 60-GHz lines [29] and optical data of A band [20] is about 7%. Our data for pure rotational lines in [7] are more close to [20], while our data of 60-GHz lines show good consistency with [29]. Some distinction in broadenings could be due to large difference in transition frequencies and experimental approaches used.

Low scattering in values of the broadenings of fine structure and rotational oxygen lines and recent optical data of oxygen electronic transitions presented in Fig. 4 allows, in our opinion, to narrow the limits of expected values of broadening parameters for unmeasured oxygen lines or could be used for evaluation of reliability of those for newly measured lines.

A good example is the case of two measurements of the rotational (N,J) = (15,14)-(13,14) transition which is of importance for remote sensing [3]. Self-broadening was measured in [31] as 2.43(43) MHz/Torr, but later in [28]—as 1.715(52) MHz/Torr by the same technique. It is easy to see that the value of [28] falls in the middle of the values plotted in Fig. 4 for the same N quantum numbers, whereas the previous value [31] lies well outside of all experimental points plotted in Fig. 4 for the same N numbers. Thus, result of [28] looks by our opinion much more realistic than that of [31].

Further, practically the same widths were observed for oxygen and nitrogen broadenings for high quantum numbers (N,J), as observed for rotational lines in [7]. The widths are even narrower for (9,9)–(7,8) line broadened by N_2 in comparison with O_2 in [28]. The same behavior is observed for the A band in [20] (see line breadths for (N,J)>15 in Table 6 of [20]). Our measurements of 7^+ , 11^+ , and 15^- lines having good SNR show unambiguous decreasing of collisional cross section in N_2 in comparison with O_2 . That is quite important for line broadening estimation in air when self-broadening data could be only available due to poorer SNR for measurements in foreign gases.

Data obtained can also be used for improvement of atmosphere propagation models and for spectroscopic databases.

4. Conclusion

The results presented in this work demonstrate a good consistency with previously obtained data by different techniques. This study has increased accuracy of the pressure broadening parameters at room temperature of 118.75 GHz oxygen line in pure gas and mixtures with N₂, Ne, He, Ar, and H₂O. To the authors' knowledge, broadening parameters by Ne, CO, and CO₂ were measured for the first time. The fine structure lines

 1^+ , 5^- , 7^+ , 11^+ , and 15^- belonging to the 60-GHz band broadened by O_2 and N_2 were also investigated. High accuracy of line center measurements has shown advantages of the RAD spectrometer for such experiments. No line shifts to be exceeded ± 10 –20 kHz/Torr were found in our investigation.

Comparison of broadenings for different types of transitions—from microwaves up to optics—shows that their values are very close in magnitude within 10% for corresponding rotational quantum number. This makes some grounds for quite reliable estimations of broadening parameters for not measured yet oxygen lines. It is worth to note that such comparison of (N,J)-dependence of Fig. 4 for three different types of oxygen spectra could be obtained only now with measurements of broadenings of a series of rotational transitions in [7] and also broadenings measurements of fine structure transitions in the present paper verifying previously measured data by another technique [29].

Acknowledgments

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