

# Microwave study of the rotational spectrum of oxygen molecule in the range up to 1.12 THz

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## Abstract

Microwave study of the rotational transitions of oxygen molecule  $^{16}\text{O}_2$  in its electronic and vibrational ground states is reported. Eight transitions belonging to  $N = 3-1$ ,  $N = 5-3$ , and  $N = 7-5$  groups were investigated. Central line frequencies and pressure broadening parameters for  $\text{O}_2$  and  $\text{N}_2$  as perturbers were determined. The highest frequency of measured transition  $(N, J) = (7, 6) - (5, 6)$  has been 1.12 THz. Spectrometer with backward wave oscillator (BWO) and acoustic detector (RAD) was used. Since this experiment has more than doubled the number of previously measured rotational lines of oxygen molecule and better accuracy was achieved, the fitting of new set of rotational transition frequencies has been performed and new more accurate molecular constants for  $^{16}\text{O}_2$  in  $X^3\Sigma_g^-, v = 0$  state have been obtained.

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## 1. Introduction

The oxygen molecule is one of the major constituents of Earth atmosphere and one of the subjects of main interest in astrophysics. Until recent times, only millimeter wave lines of fine structure of oxygen molecule were used for astrophysical and aeronautical remote-sensing purposes. Now, the rotational spectrum of oxygen molecule also attracts interest in connection with extension of airborne and satellite observational methods into submillimeter and Terahertz ranges. Rotational lines of oxygen molecule are prepared for use in airborne experiments NAST-M [1], considered for use in future in low-orbit and geostationary satellites [2], have been used in recently launched satellite ODIN [3] and submillimeter wave astronomical satellite SWAS [4], and are listed among spectroscopic needs for microwave atmosphere sensing [5]. Important parameter for the remote-sensing information is the pressure broadening of the lines by atmospheric gases. “Pure spectroscopic” information about line frequencies and molecular constants

is also of interest, having in mind very limited previous laboratory studies of the rotational spectrum of oxygen molecule. Accurate values of line frequencies are necessary for radio astronomy. Most likely causes of limited laboratory investigations are weakness of the oxygen molecular lines induced by magnetic dipole moment and difficulty of submillimeter wave spectroscopic studies.

Earlier rotational spectrum of the oxygen molecule was studied in the laboratory using microwave methods by McKnight and Gordy [6] and Steinbach and Gordy [7] who measured the frequency of one of the lowest rotational transitions  $(N, J) = (3, 2) - (1, 2)$  at 425 GHz. Pickett et al. [8] measured the broadening coefficients of the same 425 GHz line by atmospheric gases. By tunable FIR source (mix of two  $\text{CO}_2$  laser lines), Zink and Mizushima [9] measured five rotational lines of oxygen in the range from 773 GHz up to 2.5 THz frequencies. So experimental data about the rotational spectrum of oxygen molecule until the recent times consisted of six rotational transition frequencies and broadening measurements for two pure rotational lines.

As an initial stage of the rotational oxygen line study ( $^{16}\text{O}_2$ ,  $X^3\Sigma_g^-, v = 0$ ) was the reinvestigation of 425 GHz transition by Krupnov et al. [10]. In the present paper,

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we report about seven more rotational lines ranging from 368 GHz up to 1.12 THz and measurements of their central frequencies and their broadening coefficients. Spectrometer with backward wave oscillators (BWOs) and acoustic detection was used [11] and accuracy of this method was found to be significantly better than those of other methods both for line frequency and line broadening studies. Improvement of accuracy is partly due to the use of a primary radiation source (BWOs output power is 2–20 mW) but to a greater extent it is connected with the thermal character of signal detection in acoustic cell and zero baseline detection. Accuracy of measurements was verified by the control measurements of the well-known CO rotational lines within the frequency range of investigation. Internal consistency of measurements was also verified by combination analysis as comparison of differences of the measured rotational frequencies with the previously measured frequencies of the fine structure lines. Both tests confirmed the accuracy and stability of measurements reported. The more accurate than in [9] set of molecular constants of oxygen molecule  $^{16}\text{O}_2$  in the ground state was obtained, as a result of the processing of the new data with the known frequencies. Accurate measurements of line broadenings showed a tendency of decreasing of the line broadening with the increase of rotational quantum number in agreement with infrared [12] and optical studies [13].

## 2. Experiment

For the study of the rotational spectrum of oxygen in the submillimeter wave range, the RAD spectrometer with BWOs and the acoustic cell [11] was employed as in [10]. The block-scheme of the experimental setup is the same as that depicted in Fig. 1 of [10], with the only difference being that in the present study additional BWO sources in the range 258–375 GHz (OB-30 or OB-65), 667–857 GHz (OB-81), 789–968 GHz (OB-82), and 1034–1250 GHz (OB-84) were used. Frequency of BWOs sources was phase locked against harmonics of fundamental output frequency of 78–118 GHz “KVARZ” synthesizer of type [14]. The synthesizer frequency itself was stabilized against Rubidium standard. Details of such spectrometer were given in [10,15].

As in [10] for the study of the oxygen transition sensitive to ambient magnetic field, the double magnetic shell was used. The shell was made of annealed perm-alloy to shield the cell with the oxygen sample. The small size of the acoustic cell ( $\sim 10$  cm length,  $\sim 2$  cm diameter) permitted a convenient and compact shielding construction. The gas pressure in the cell was measured by MKS Baratron gauge. Pure gas samples from Messer MG Company were used in the investigations.

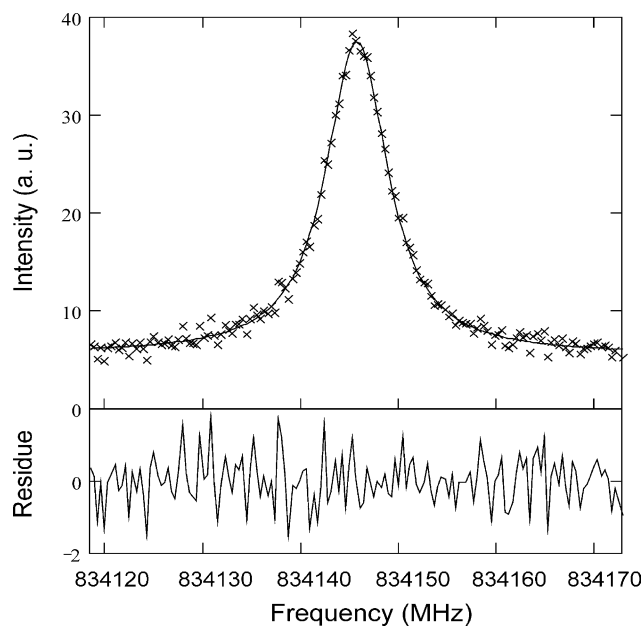


Fig. 1. Record of the  $(N, J) = (5, 5) - (3, 4)$  rotational line of the oxygen molecule. Experimental points are denoted by cross ( $\times$ ), the line (—) represents the fit to Voigt line profile; bottom trace is residue of the fit. Pressure in the cell was  $P = 1.81$  Torr.

Records of the typical lines with fittings to Voigt profile model and the residue of the fit are presented in Figs. 1 and 2. The line in Fig. 1 belongs to the  $N = 5-3$  group, while the line in Fig. 2 belongs to the  $N = 7-5$  group and has the highest frequency measured about 1.12 THz. The record of the line belonging to the  $N = 3-1$  group is shown in Fig. 2 of [10].

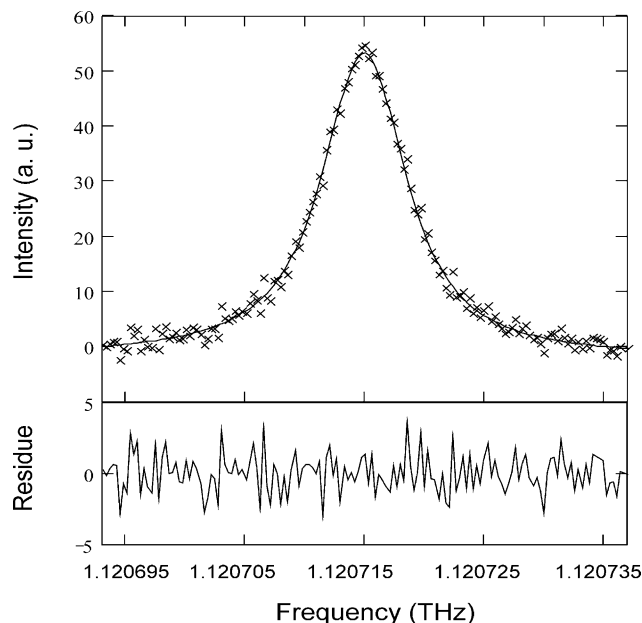


Fig. 2. The  $(N, J) = (7, 6) - (5, 6)$  of  $^{16}\text{O}_2$  rotational line. Transition frequency is 1.12071484(5) THz, gas pressure was  $P = 1.93$  Torr at  $T = 302$  K.

### 3. Results of experiment

The measured frequencies and broadening parameters are collected in Table 1. The data on 425 GHz transition [10] are also included. Frequencies of the lines were obtained as a result of the fit of line shapes to Voigt profile (Figs. 1 and 2). No significant pressure line shifts were found (to have exceeded 100 kHz/Torr, as it was mentioned in [10]). The accuracy and stability of the line frequency measurements were checked by the control measurements of the rotational lines of CO molecule used as secondary calibration standard. Measurements of CO lines were done throughout the entire frequency range studied. In Table 2, values of our measured frequencies of CO lines are listed together with calculated frequencies of CO lines obtained from a fit of all precisely measured lines of its rotational spectrum [16]. Differences between measured and calculated lines lie well within the errors and vary from 0.55 kHz for the lowest  $J = 3 \leftarrow 2$  transition at 345 GHz to the 6.8 kHz for the highest frequency measured by us  $J = 9 \leftarrow 8$  transition at 1 036 GHz. These values demonstrate the limits of the possible systematic errors of the RAD spectrometer.

The internal consistency of frequency measurements was also checked by the fact that the oxygen molecule rotational levels have a magnetic fine structure, transitions of which were well measured in millimeter wave band (see e.g., [9,17]). This fact permitted us to compare from one side—differences between frequencies of rotational lines measured by us, and from another side—frequencies of the fine structure lines, which mostly fall at frequencies around 60 GHz [17]. As it can be seen in Table 3, agreement between differences of frequencies of the rotational lines measured in this work and frequencies of corresponding fine structure lines is better than the quoted errors.

The collisional line half-widths at half-maximum height (HWHM) were defined from the fit of the experimental line shapes to the Voigt profile (as it is shown in Figs. 1 and 2). The calculated values of Doppler linewidths at room temperature were used in the fit. Example of the linear regression fit of collisional HWHM (MHz) values versus oxygen pressure (Torr) together with the residue of this fit is presented in Fig. 3 of [10]. The broadening parameter was then defined from the slope of linewidth dependence on pressure.

Table 1

Experimental line frequencies and broadening parameters for  $^{16}\text{O}_2$  ( $X^3\Sigma_g^-, v = 0$ ) rotational transitions

Transition $N', J' \leftarrow N, J$	Observed frequency (MHz)	Broadening parameters (MHz/Torr) by $\text{O}_2$	Broadening parameters (MHz/Torr) by $\text{N}_2$
3, 2–1, 1	368 498.245 (0.02)	$2.21 \pm 0.03$ (31 °C)	—
3, 2–1, 2	424 763.023 (0.02)	$2.19 \pm 0.01$ (23 °C)	$2.215 \pm 0.02$ (23 °C)
3, 3–1, 2	487 249.27 (0.03)	$2.10 \pm 0.03$ (24 °C)	$2.16 \pm 0.1$ (26 °C)
5, 4–3, 3	715 392.98 (0.07)	$2.08 \pm 0.1$ (27 °C)	—
5, 4–3, 4	773 839.51 (0.06)	$2.01 \pm 0.06$ (26.5 °C)	$2.2 \pm 0.18$ (26.5 °C)
5, 5–3, 4	834 145.560 (0.05)	$1.94 \pm 0.07$ (28 °C)	$1.96 \pm 0.15$ (28.5 °C)
7, 6–5, 5	1 061 123.857 (0.1)	$1.94 \pm 0.12$ (29.5 °C)	—
7, 6–5, 6	1 120 714.836 (0.04)	$1.94 \pm 0.06$ (28.5 °C)	$1.98 \pm 0.15$ (29 °C)

Table 2

The frequencies of the rotational CO lines measured in this work and calculated in [16]

Transition, $J' \leftarrow J$	Frequency (MHz), this work meas.	Frequency (MHz), calc. from [19]	Obs. – calc. (kHz)
3–2	345 795.9904 (0.001)	345 795.98985 (0.16 kHz)	0.55
4–3	461 040.7725 (0.005)	461 040.76798 (0.21 kHz)	4.5
5–4	576 267.933 (0.005) <sup>a</sup>	576 267.93101 (0.25 kHz)	2
7–6	806 651.80 (0.010)	806 651.8010 (0.3 kHz)	–1
9–8	1 036 912.392 (0.009)	1 036 912.3852 (0.5 kHz)	6.8

<sup>a</sup> From [15].

Table 3

Frequencies of fine structure transitions obtained as differences of measured by us rotational frequencies and previously measured fine structure lines

Loops and transitions	Calculated from this experiment (MHz)	Experimental values given in [17] (MHz)
(3, 2–1, 2)–(3, 2–1, 1) = (1, 1–1, 2)	56 264.778 (40)	56 264.772 (10)
(3, 3–1, 2)–(3, 2–1, 2) = (3, 3–3, 2)	62 486.247 (30)	62 486.255 (10)
(5, 4–3, 4)–(5, 4–3, 3) = (3, 3–3, 4)	58 446.530 (70)	58 446.585 (10)
(5, 5–3, 4)–(5, 4–3, 4) = (5, 5–5, 4)	60 306.050 (60)	60 306.044 (20)
(7, 6–5, 6)–(7, 6–5, 5) = (5, 5–5, 6)	59 590.979 (100)	59 590.978 (20)

The newly obtained and earlier known frequencies of rotational and fine structure lines were fitted all together by JPL program *spfit.exe* [18] to the same set of molecular constants as in [9]. The set of transitions used, experimental and calculated frequencies and differences between them are listed in Table 4.

In the fit procedure, we used previously obtained experimental data together with the ones measured in this work and having the same weight. But inspection of Table 4 shows that the rotational lines measured in the present study are more self-consistent and more close to prediction than the rotational lines measured in [7] and [9]. That gave us the motivation to make another fit.

So in Table 5 the fit is presented, in which we used only the rotational frequencies measured in this work together with the known fine structure frequencies [17]. For brevity, only the part with the rotational lines is presented in Table 5 (fitted values of fine structure lines were practically the same as given in Table 4). As it can be seen from Table 5, the discrepancies between measured and calculated values become much lesser than those given in Table 4 what demonstrates good internal consistency of the present measurements.

In Table 6, the set of molecular constants of oxygen molecule  $^{16}\text{O}_2$  ( $X^3\Sigma_g^-, v=0$ ) obtained from the fit of all known frequencies and newly obtained ones is

Table 4  
Observed and calculated oxygen  $^{16}\text{O}_2$  lines ( $X^3\Sigma_g^-, v=0$ )

Transition				Observed frequency (MHz)	Calculated frequency (MHz)	Obs. – calc. (MHz)
$N'$	$J'$	$N$	$J$			
21	21	21	20	54 671.145(20) [17]	54 671.16543	–0.02043
19	19	19	18	55 221.372(10) [17]	55 221.37123	0.00077
17	17	17	16	55 783.819(20) [17]	55 783.80387	0.01513
1	1	1	2	56 264.772(10) [17]	56 264.77624	–0.00424
15	15	15	14	56 363.393(10) [17]	56 363.38949	0.00351
13	13	13	12	56 968.180(20) [17]	56 968.20435	–0.02435
9	9	9	8	58 323.885(20) [17]	58 323.87364	0.01136
3	3	3	4	58 446.585(10) [17]	58 446.59059	–0.00559
7	7	7	6	59 164.215(20) [17]	59 164.20272	0.01228
5	5	5	6	59 590.978(10) [17]	59 590.98486	–0.00686
5	5	5	4	60 306.044(20) [17]	60 306.05647	–0.01247
7	7	7	8	60 434.776(10) [17]	60 434.77929	–0.00329
9	9	9	10	61 150.570(10) [17]	61 150.56285	0.00715
11	11	11	12	61 800.169(20) [17]	61 800.15765	0.01135
13	13	13	14	62 411.223(10) [17]	62 411.21880	0.00420
3	3	3	2	62 486.255(10) [17]	62 486.25445	0.00055
17	17	17	18	63 568.520(10) [17]	63 568.52145	–0.00145
19	19	19	20	64 127.777(20) [17]	64 127.76937	0.00763
25	25	25	26	65 764.744(20) [17]	65 764.77100	–0.02700
31	31	31	30	52 021.423(09) [17]	52 021.42094	0.00206
33	33	33	32	51 503.348(18) [17]	51 503.35815	–0.01015
35	35	35	34	50 987.760(19) [17]	50 987.75051	0.00949
37	37	37	36	50 474.223(27) [17]	50 474.23091	–0.00791
33	33	33	34	67 900.867(11) [17]	67 900.86267	0.00433
35	35	35	36	68 431.005(17) [17]	68 431.00294	0.00206
37	37	37	38	68 960.312(12) [17]	68 960.31235	–0.00035
39	39	39	40	69 489.030(15) [17]	69 489.03247	–0.00247
1	1	1	0	118 750.343(10) [6]	118 750.33803	0.00497
3	2	1	1	368 498.245(20) <sup>a</sup>	368 498.25900	–0.01400
3	2	1	2	424 763.023(20) <sup>a</sup>	424 763.03523	–0.01223
3	2	1	2	424 763.210(100) [7]	424 763.03523	0.17477
3	3	1	2	487 249.270(30) <sup>a</sup>	487 249.28968	–0.01968
5	4	3	3	715 392.980(70) <sup>a</sup>	715 392.92969	0.05031
5	4	3	4	773 839.691(64) [9]	773 839.51994	0.17106
5	4	3	4	773 839.510(60) <sup>a</sup>	773 839.51994	–0.00994
5	5	3	4	834 145.560(50) <sup>a</sup>	834 145.57676	–0.01676
7	6	5	5	1 061 123.857(100) <sup>a</sup>	1 061 123.87667	–0.01967
7	6	5	6	1 120 714.836(50) <sup>a</sup>	1 120 714.86153	–0.02553
9	8	7	8	1 466 807.133(60) [9]	1 466 806.97455	0.15845
11	10	9	10	1 812 405.539(112) [9]	1 812 405.31216	0.22684
13	12	11	12	2 157 577.773(52) [9]	2 157 577.79317	–0.02017
15	14	13	14	2 502 323.923(92) [9]	2 502 324.00223	–0.07923

The frequencies from [6,7,9,17] and from the present experiment are used in the fit.

<sup>a</sup> Present work.

Table 5

Observed and calculated oxygen  $^{16}\text{O}_2$  lines ( $X^3\Sigma_g^-, v = 0$ )

$N'$	$J'$	$N$	$J$	Observed frequency (MHz)	Calculated frequency (MHz)	Obs. – calc. (MHz)
1	1	1	0	118 750.343(10) <sup>a</sup>	118 750.33804	0.00496
3	2	1	2	368 498.245(20)	368 498.24754	–0.00254
3	2	1	2	424 763.023(20)	424 763.02394	–0.00094
3	3	3	3	487 249.270(30)	487 249.27823	–0.00823
5	4	3	4	715 392.980(70)	715 392.91262	0.06738
5	4	3	4	773 839.510(60)	773 839.50320	0.00680
5	5	5	5	834 145.560(50)	834 145.55968	0.00032
7	6	5	6	1 061 123.857(100)	1 061 123.86063	–0.00363
7	6	5	6	1 120 714.836(50)	1 120 714.84539	–0.00939

The rotational frequencies from the present work only are used in the fit.

<sup>a</sup>One of fine structure transitions as an example taken from [6].

Table 6

Molecular constants of oxygen  $^{16}\text{O}_2$  in the  $X^3\Sigma_g^-, v = 0$  state obtained from the fit of all known lines in the ground state in comparison to the values reported in [9]

Constant	This work (MHz)	Ref. [9] (MHz)
$B$	43 100.4438 (9)	43 100.4519 (23)
$D$	0.145115 (40)	0.14514 (78)
$\lambda$	59 501.3459 (33)	59 501.346 (21)
$\lambda_D$	0.058326 (21)	0.058309(61)
$\lambda_{DD}$	$3.240(17) \times 10^{-7}$	$3.34(146) \times 10^{-7}$
$\gamma$	–252.58653 (45)	–252.5867 (21)
$\gamma_D$	$-2.435(13) \times 10^{-4}$	$-2.45296(175) \times 10^{-4}$
$\gamma_{DD}$	$-1.14(83) \times 10^{-9}$	$-1.44(30) \times 10^{-9}$

presented. For comparison, the set of previously obtained constants in [9] is also given in Table 6.

#### 4. Discussion

The new set of molecular constants of oxygen molecule shows the better accuracy of the most part of constants. A significant improvement of accuracy was achieved for rotational and centrifugal constants  $B$  and  $D$ , and for spin–spin interaction constants  $\lambda$ ,  $\lambda_D$ ,  $\lambda_{DD}$  and spin–rotation constant  $\gamma$  as it follows from Table 6. In contrast with that, the values of the other spin–rotation constants  $\gamma_D$  and  $\gamma_{DD}$  were found as defined to be somewhat poorer than in [9].

The results in Table 1 show uniform and smooth behavior of the values of both self-broadening and nitrogen-broadening parameters for the rotational lines investigated. Clear tendency to the decreasing of both the (self- and nitrogen-) broadening parameters with the increase of the rotational quantum number can be noticed. For the self-broadening parameter, a value of 2.21 MHz/Torr for  $N = 3$ –1 line at 368 GHz decreases to 1.94 MHz/Torr for the line  $N = 7$ –5 at 1.12 THz. For the nitrogen-broadening parameter value 2.215 MHz/Torr for  $N = 3$ –1 line at 425 GHz decreases to 1.98 MHz/Torr for the line  $N = 7$ –5 at 1.12 THz.

The pressure self-broadening parameter 2.006(64) MHz/Torr ( $T = 290$  K) quoted in [19] for submillimeter

transition  $J = 7 \leftarrow 6$  of a  $^1\Delta_g\text{O}_2$  state and values obtained in [12] 1.93–2.13 MHz/Torr for vibration–rotation lines of the  $a^1\Delta_g-X^3\Sigma_g^-$  band of about the same  $J$ -numbers and values obtained in this work look to be consistent.

It is also interesting to compare our broadening parameters of rotational lines with values of electronic A band transitions measured in [13]. Comparison shows similarity both in values of broadenings and in their dependences on rotational quantum numbers. Indeed, for the self-broadening parameter values ranging from 2.32 MHz/Torr to 2.035 MHz/Torr are reported in [13] for low  $J$ -transition range (P1P1–P5P5); these values decrease to 1.86 MHz/Torr self-broadening parameter for R9R9. For nitrogen broadening, corresponding values are from 2.34 MHz/Torr to 2.07 MHz/Torr for P1P1–P5P5 transitions and decrease to 1.87 MHz/Torr for R9R9.

Broadening values and dependencies on quantum numbers of the other types of transition of oxygen molecule [12,13] cannot be used for immediate prediction of the value of broadenings of, e.g., rotational transition  $N = 15$ –13 at 2.502 THz, which is of interest for future experiments on remote sensing [5]. But joint consideration of the results of [12,13] and of the present study permitted by our opinion some guessing about possible limits of these transition broadenings. In view of the aforementioned 2.502-THz line ( $N = 15$ –13), the value of the self-broadening parameter measured in [9] as 2.43(43) MHz/Torr looks rather as an upper limit.

The pressure-induced line shifts of the order of—(100–200) kHz/Torr were measured in [13] for A band transitions. As it was said, we did not observe the pressure shifts for the rotational lines in the limits mentioned above and plan to return to these studies in the future.

#### 5. Conclusion

To the authors' knowledge, the present investigation represents the most complete and accurate study of

rotational spectrum of the oxygen molecule since [9]. The experiment has more than doubled the number of measured rotational lines of the oxygen molecule and more accurate set of molecular constants has been obtained.

Broadenings of seven more rotational lines were measured. Tendency of decreasing of the broadening with the increase of rotational quantum number is found in agreement with infrared and optical studies.

Spectroscopic needs for remote sensing in [5] listed the broadenings of the two rotational lines of oxygen molecule at 487 GHz and at 2.502 THz. In the present study, the broadenings of the first line are directly measured. And based on values of consecutive rotational transitions from 368 GHz up to 1.12 THz, some estimation was given for broadenings of the second line. So from our point of view the broadening parameters of 2.502 THz line are worth being reinvestigated with better accuracy.

Data obtained can be used also for improvement of atmosphere propagation models [20].

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