

CONCERNING THE WIDTHS AND INTENSITIES OF  
SUBMILLIMETER ABSORPTION LINES OF THE  
ROTATIONAL SPECTRUM OF WATER VAPOR

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UDC 621.371.166.2

Experimental data are presented on the absorption coefficient of water vapor in the resonance regions of the lines corresponding to the  $1_{-1} - 1_1$  ( $\lambda_{ij} = 0.538$  mm) and  $2_2 - 3_{-2}$  ( $\lambda_{ij} = 1.635$  mm) transitions. The measurements were carried out in atmospheric air using backward-wave tubes and pneumatic OAP-2 receivers. The measured contours of the absorption lines are used to determine their widths, intensities, and resonance wavelengths. A summary of the experimental values of the half-width and integral intensity (forces) of the rotational  $H_2O$  line obtained by various authors in the  $\lambda > 0.05$  mm range is given, and they are compared with each other and with theoretical computations.

Water vapor, due to the fact that it has a complicated and very extended (from infrared to centimeter wavelengths) rotational spectrum, plays an important role in various physical phenomena which go on in the Earth's atmosphere — such as molecular absorption, refraction and fluctuation of microradiowaves, radiative heat exchange, etc. This fact aroused the interest which has already been manifested for approximately four decades by various researchers in the theoretical and experimental study of the structure of the rotational spectrum of  $H_2O$  vapors.

By now, the identification of spectral lines has been mainly completed as a result of numerous spectroscopic investigations covering essentially the entire range occupied by the rotational spectrum of  $H_2O$ . Fairly complete and exact data on the resonance frequencies of the lines [1-7] and the energy levels of a water molecule [5, 8-10] have been obtained.

The widths and intensities of the lines have been studied to a considerably lesser degree. Theoretically, the width of the rotational lines of water vapor for the main types of broadening molecular interactions in the atmosphere ( $H_2O - N_2$ ,  $H_2O - H_2O$ , and  $H_2O - O_2$ ) were calculated by Benedict and Kaplan [11, 12]. However, without a reliable experimental check of these calculations it is difficult to evaluate the extent to which the values of the widths obtained in [11, 12] are close to the true values.

As far as the intensities of the spectral lines of  $H_2O$  are concerned, it follows that here the theoretical data are based on computations [13] of the matrix elements of the directional cosines for the rigid-asymmetrical-top model. The estimates of the corrections due to centrifugal perturbation of the  $H_2O$  molecule have been performed in [6, 14, 15].

Systematic experimental investigations of the widths and intensities of  $H_2O$  lines have not been carried out until recently due to the absence of sufficiently sensitive and highly rotational spectral equipment. Measurements were performed of the parameters of the microwave line  $\lambda_{ij} = 1.35$  cm [16-18] and of the width of two lines in the infrared range ( $\lambda_{ij} = 15.99$  and  $18.64$   $\mu$ ) [19].

During the last 10 years the situation has changed noticeably. Due to the successful advance of radio and optics methods in the submillimeter range and the development of new types of instruments, microwave-spectrometer measurements have been carried out of the parameters of strong longwave lines  $\lambda_{ij} = 1.635$  mm [20-27],  $\lambda_{ij} = 0.922$  mm [27-30],  $\lambda_{ij} = 0.788$  mm [27, 31, 32],  $\lambda_{ij} = 0.538$  mm [24],  $\lambda_{ij} = 0.398$  mm [33], as well as spectrometric investigations [34-36] of the most isolated lines in the 0.05 to 0.6 mm

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Scientific-Research Institute of Radiophysics. Translated from *Izvestiya Vysshikh Uchebnykh Zavedenii, Radiofizika*, Vol. 18, No. 3, pp. 358-370, March, 1975. Original article submitted June 13, 1973.

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range ( $\lambda_{ij} = 53.1$  and  $58.7 \mu$  [34],  $\lambda_{ij} = 0.108$  mm [34, 36],  $\lambda_{ij} = 0.212$  and  $0.398$  mm [36], and  $\lambda_{ij} = 0.538$  mm [35, 36]). The methods of optical spectroscopy have likewise been used to investigate a series of lines of the rotational spectrum of  $H_2O$  in the infrared range – 19 lines in the  $\lambda = 14.5$ – $21 \mu$  sector of the spectrum [6] and the  $\lambda_{ij} = 10.94 \mu$  line [37]. Finally, the parameters of the microwave line  $\lambda_{ij} = 1.35$  cm [38–40] have been studied in greater detail.

The present paper pursues two aims. First, we reworked the data of our measurements performed in [24] which were cited in the review [14]. Now, due to the use of a more correct method of handling which takes account of the self-broadening of the lines and eliminates the systematic errors caused by temperature and pressure variations during the measurements, the results of these measurements have been refined. Below, a handling procedure is expounded, and a previously unpublished description of the measurements themselves is likewise presented.

Second, since a significant amount of experimental material has already been accumulated on the widths and intensities of the lines of the rotational spectrum of  $H_2O$ , it seemed expedient to perform a detailed comparison between available experimental data both with each other and with the results of theoretical computations.\* Such a comparison is performed below for the range of wavelengths  $\lambda > 50 \mu$ .

#### Measurement of the Parameters of the $\lambda_{ij} = 0.538$ mm Line

The absorption line  $\lambda_{ij} = 0.538$  mm (the transition  $1_{-1} - 1_1$ ) belongs among those strong and isolated lines in whose resonance region the background of the wings of the remaining lines is negligibly small. This allows a line contour undistorted by background to be obtained from experiments even at atmospheric air pressure.

In order to obtain the contour of the line, measurements of the absorption coefficient of atmospheric water vapor were performed at a series of wavelengths in the  $530$ – $545 \mu$  interval. The required spectral resolution was ensured by using a source of monochromatic radiation (a backward-wave tube) [41]. The reception of signals is accomplished by means of an OAP-2 pneumatic detector. In the high-frequency portion of the installation an off-axial elliptical mirror was used whose foci were made to coincide with the phase center of the horn radiator of the backward-wave tube and the receiving area of the OAP-2. The optical path in air had a length  $L = 0.9$  m.

The absorption measurements were performed by the method of variation of the absolute air humidity. Since the experiment was carried out in the summertime, the lowest value of humidity was  $\sim 10$  g/m<sup>3</sup>. The air humidity was raised artificially to  $\rho = 21$ – $23$  g/m<sup>3</sup>. For such a humidity variation, the chosen optical path length (0.9 m) ensures significant changes of the signals with humidity at all frequencies (at the resonant frequency of the line this change was by more than a factor of 100) and at the same time a completely reliable recording of it in the peak of the absorption line.

The measurements were carried out as follows. First, the signal level was recorded successively at each of the chosen wavelengths at the highest air humidity. Then a rapid transition was effected to the lowest humidity corresponding to the humidity of the outside air, and the signal level was measured at the same wavelength. The merit of such a "two-point" method resides in the fact that in order to perform one complete cycle of measurement one requires a comparatively short time (less than 1 h), as a consequence of which the effect of various kinds of equipment instabilities turned out to be of little significance.

The wavelength was measured by a wavemeter of the Fabry – Perot interferometer type. The accuracy of the measurements was  $\sim \pm 0.02\%$ .

Let us dwell on the data-handling method. When measurements are performed by the method of variation of the air humidity, the signal reduction is determined not only by the variation of the mass of absorbing gas  $\rho L$  but to a certain extent also by the fact that due to self-broadening of the spectral line the magnitude of the "specific" (for 1 g/m<sup>3</sup> of  $H_2O$  vapors) absorption coefficient  $\gamma$  of the water vapor also varies when  $\rho$  varies. Moreover, since  $\gamma$  depends on the temperature  $T$  and the pressure  $P$  of dry air, the variation of these parameters which usually accompany the measurements also affects the signal level. The character of the dependence of  $\gamma$  on both  $\rho$  and on  $T$  and  $P$  in the region of the resonance of the line varies with wavelength. In principle, these dependences may be established theoretically for each wavelength, and then one can use them to take account of the effect of  $\rho$ ,  $T$ ,  $P$  on the magnitude of  $\gamma$  in handling the experimental

\*The review table of linewidths which was published in 1967 in [14] currently needs both a series of refinements and natural supplementation as a result of the data obtained from measurements performed in recent years.

material. The shortcoming of such a method, besides its cumbersome nature, lies in the fact that the inaccuracy of the theoretical functions  $\gamma_\lambda(\rho)$ ,  $\gamma_\lambda(T)$ ,  $\gamma_\lambda(P)$  used may lead to the appearance of systematic errors. Specifically, the measurements performed in [42] yielded a temperature dependence differing from the theoretical one.

It is therefore preferable to use a different method of handling which does not require the use of specific dependences of  $\gamma$  on  $\rho_1$ ,  $T$ ,  $P$ . It consists in the following. Let the state of the absorptive air be characterized by the parameters  $\rho_1$ ,  $T_1$ ,  $P_1$  for the lowest humidity and by the parameters  $\rho_2$ ,  $T_2$ ,  $P_2$  for the highest humidity. Then on the basis of the Bouguer – Beer law for the intensities  $I_1$  and  $I_2$  of the measured signals in these states, we have

$$\ln I_1 = \ln I_0 - \gamma(\rho_1, T_1, P_1) \rho_1 L; \quad (1a)$$

$$\ln I_2 = \ln I_0 - \gamma(\rho_2, T_2, P_2) \rho_2 L, \quad (1b)$$

where  $I_0$  is the intensity in the absence of absorption. Subtracting (1b) from (1a) and expanding the function  $\gamma$  into a series in the neighborhood of certain values of  $\rho_0$ ,  $T_0$ ,  $P_0$ , we obtain

$$\begin{aligned} \frac{\ln I_1 - \ln I_2}{L} = & \gamma(\rho_0, T_0, P_0) (\rho_2 - \rho_1) + \left(\frac{\partial \gamma}{\partial \rho}\right)_0 [(\rho_2 - \rho_0) \rho_2 - (\rho_1 - \rho_0) \rho_1] \\ & + \left(\frac{\partial \gamma}{\partial T}\right)_0 [(T_2 - T_0) \rho_2 - (T_1 - T_0) \rho_1] + \left(\frac{\partial \gamma}{\partial P}\right)_0 [(P_2 - P_0) \rho_2 - (P_1 - P_0) \rho_1]. \end{aligned} \quad (2)$$

In view of the comparatively weak dependence of  $\gamma$  on  $\rho$  and the smallness of the relative variations of  $T$  and  $P$ , only linear terms of the expansion are taken into account in (2).

If the expansion point is taken in such a way that

$$\begin{aligned} \rho_0 = \rho^* = \rho_1 + \rho_2, \\ T_0 = T^* = \frac{T_2 \rho_2 - T_1 \rho_1}{\rho_2 - \rho_1}, \quad P_0 = P^* = \frac{P_2 \rho_2 - P_1 \rho_1}{\rho_2 - \rho_1}, \end{aligned} \quad (3)$$

then in accordance with (2) we have

$$\gamma(\rho^*, T^*, P^*) = \frac{1}{L} \frac{\ln I_1 - \ln I_2}{\rho_2 - \rho_1}. \quad (4)$$

Thus, for measurement of absorption at some wavelength, the effect of self-broadening and variations of  $T$  and  $P$  can easily be taken into account by introducing the parameters  $\rho^*$ ,  $T^*$ ,  $P^*$ . In order to obtain an undistorted contour of the line, the values of these parameters for all wavelengths must be appropriately close to one another.

In the present experiment, several series of measurements were performed for which  $\rho^* = 32 (\pm 0.5)$  g/m<sup>3</sup>,  $T^* = 303 (\pm 1.5)$  K,  $P^* = 715 (\pm 3)$  mm Hg. Figure 1 displays the experimental absorption coefficients of water vapor for these values of  $\rho^*$ ,  $T^*$ ,  $P^*$ . The mean-square error of the data shown in Fig. 1, which is determined according to the spread of the results obtained in individual series of measurements, amounts to  $\sim \pm 2\%$  at frequencies close to the resonance of the line and  $\sim \pm 5\%$  in its wings.

In order to determine the parameters of the line [the resonant frequency  $\nu_{ij}$ , the half-width  $\Delta\nu_{ij}$ , and the absorption coefficient in the peak of the line  $\gamma_{ij}(\nu_{ij})$ ], an expression was used which describes the shape of the isolated spectral line [14]:

$$\gamma_{ij}(\nu) = \frac{4\gamma_{ij}(\nu_{ij}) \nu_{ij}^2 (\Delta\nu_{ij})^2}{(\nu^2 - \nu_{ij}^2)^2 + 4\nu^2 (\Delta\nu_{ij})^2}. \quad (5)$$

By choosing the parameters  $\nu_{ij}$ ,  $\Delta\nu_{ij}$ , and  $\gamma_{ij}(\nu_{ij})$  in (5), one can find a curve which best approximates the spectral distribution  $\gamma^{\text{exp}}(\nu)$  obtained in the experiment (Fig. 1). Under these conditions the contribution which the wing of the other lines make to the absorption is neglected [according to estimates, this contribution is only 0.3% of the value of  $\gamma^{\text{exp}}(\nu_{ij})$ ].

The parameters of the line which were found in this manner and correspond to the values  $\rho^*$ ,  $T^*$ ,  $P^*$  indicated above turned out to equal

$$\begin{aligned} \nu_{ij}^{\text{exp}} = 18.579 \pm 0.003 \text{ cm}^{-1}, \quad \Delta\nu_{ij}^{\text{exp}} = 0.116 \pm 0.004 \text{ cm}^{-1}, \\ \gamma_{ij}^{\text{exp}}(\nu_{ij}) = 2050 \pm 40 \text{ dB} \cdot \text{km}^{-1} / \text{g} \cdot \text{m}^{-3}. \end{aligned}$$

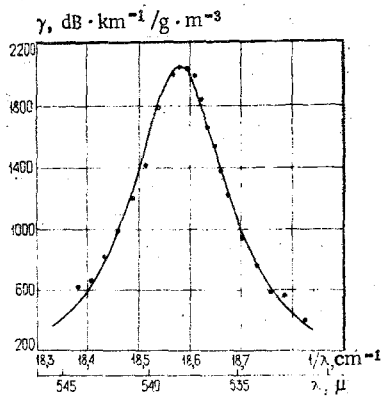


Fig. 1

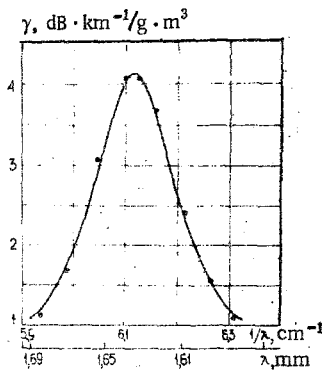


Fig. 2

Fig. 1. Experimental contour of the spectral line  $\lambda_{ij} = 0.538$  mm. The points are the measured values of the absorption coefficient of  $H_2O$  vapors; the solid line is the approximating curve.

Fig. 2. Results of absorption measurements in the region of resonance with the  $\lambda_{ij} = 1.635$  mm line. The solid line is the approximating curve.

In estimating the errors of the results, the accuracy with which the wavelength was measured, the mean-square deviation of the points from the approximating curve, and the results of comparing the behavior of the experimental points with the curves (5) plotted for various values of the line parameter were taken into account.

#### Measurement of the Parameters of the $\lambda_{ij} = 1.635$ mm Line

The absorption line  $\lambda_{ij} = 1.635$  mm is considerably lower in intensity than the line  $\lambda_{ij} = 0.538$  mm considered above; therefore, measurements of the absorption in its resonance region require the use of a long optical path. In our measurements, the distance between the transmitter and receiver amounted to 1.58 km. The measurements were carried out in the fall of 1966 under field conditions by the varying-humidity method. The equipment and procedure used for the measurements were analogous to those described in [43].

In handling the data, the self-broadening of the line was taken into account by introducing the humidity  $\rho^*$  as in the case of two-point measurements, and here too we have  $\rho^* = \rho_1 + \rho_2$ . As far as considering the changes in temperature and pressure is concerned, the most important thing is to eliminate the systematic errors of  $T(\rho)$  and  $P(\rho)$ . It may be shown that when the behavior of  $T(\rho)$  and  $P(\rho)$  is almost linear,† the systematic errors will be eliminated if the results of the measurements are referred to  $T^* = T(\rho^*)$  and  $P^* = P(\rho^*)$ . In the given experiment  $\rho_1 \approx 3 \text{ g/m}^3$ ,  $\rho_2 \approx 7 \text{ g/m}^3$ , and, consequently,  $\rho^* \approx 10 \text{ g/m}^3$ . The variations of temperature and pressure were satisfactorily approximated by the dependence of  $T = 272 + 1.7(\rho - \rho_1)$ ,  $P = 759 - 4.2(\rho - \rho_1)$  so that  $T^* \approx 284^\circ\text{K}$  and  $P^* \approx 730 \text{ mm Hg}$ .

The measured results corresponding to these values  $\rho^*$ ,  $T^*$ ,  $P^*$  are displayed in Fig. 2. The mean-square errors of the measured values of  $\gamma$  amount to 4-6%.

In determining the parameters of the line we took into account the fact that the spectral distribution  $\gamma^{\text{exp}}(\nu)$  obtained from the measurements constitutes the sum

$$\gamma^{\text{exp}}(\nu) = \gamma_{ij}(\nu) + \gamma_b(\nu), \quad (6)$$

whose first term describes the absorption which is associated solely with the investigated  $ij$ -transition [viz., (5)], while the second takes account of the contribution (the "background") from the wings of all the

†Such a case is frequently realized in measurements performed under field conditions when the growth in humidity is associated with an increase in temperature, while the atmospheric pressure  $P_{\text{atm}}$  generally varies little and on the average may usually be assumed constant, since the pressure variation of dry air  $P = P_{\text{atm}} - P_{H_2O}$  may be described approximately by a linear function of  $\rho$ .

remaining lines. In order to determine  $\gamma_b$ , the experimental data in [23, 44] on absorption in the transparency windows adjacent to the considered line were used. According to estimates,  $\gamma_b \approx 0.2 \text{ dB} \cdot \text{km}^{-1} / \text{g} \cdot \text{m}^{-3}$  in the resonance region of the line.

The choice of parameters in (6) in seeking the curve which approximates the behavior of the point in Fig. 2 led (for  $\rho^* = 10 \text{ g/m}^3$ ,  $T^* = 284^\circ\text{K}$ ,  $P^* = 730 \text{ mm Hg}$ ) to the values

$$\begin{aligned} \nu_{ij}^{\text{exp}} &= 6.117 \pm 0.005 \text{ cm}^{-1}, \quad \Delta\nu_{ij}^{\text{exp}} = 0.105 \pm 0.006 \text{ cm}^{-1}, \\ \gamma_{ij}^{\text{exp}}(\nu_{ij}) &= 3.9 \pm 0.2 \text{ dB} \cdot \text{km}^{-1} / \text{g} \cdot \text{m}^{-3}. \end{aligned}$$

The errors of the measurements were estimated by the same method as that used for the  $\lambda_{ij} = 0.538 \text{ mm}$  line. The possible error due to the inaccuracy of the value of  $\gamma_b$  was additionally taken into account.

## Discussion of the Results of the Measurements. Brief

### Review of Experimental Data and Comparison with Theory

The Widths of the Lines. The comparison of the experimental half-widths of the  $\lambda_{ij} = 0.538 \text{ mm}$  and  $\lambda_{ij} = 1.635 \text{ mm}$  lines with results of other measurements and computations [11, 12] is made in Table 1. This table likewise includes all the experimentation known to us (with the exception of data having a low accuracy which were reported in [16, 20, 45]) on the widths of other submillimeter lines ( $\lambda_{ij} > 50 \mu$ ) and of the microwave line  $\lambda_{ij} = 1.35 \text{ cm}$ .†

Note that in a number of the papers cited (in [23, 25, 28, 29, 31-33] and partially in [27]), as well as in the present measurements, the values found for the half-widths apply to wet gases (i.e., in general, they are determined by two types of molecular collisions:  $\text{H}_2\text{O}$  and dry expanding gas, and  $\text{H}_2\text{O}-\text{H}_2\text{O}$  involving self-broadening of the line). Due to the differences in the humidities of the gas, the values of  $\Delta\nu_{ij}^{\text{exp}}$  obtained in different experiments obviously contain a nonidentical contribution caused by  $\text{H}_2\text{O}-\text{H}_2\text{O}$  collisions; this makes comparison of available data somewhat difficult. In view of this, the contribution of self-broadening (from 3 to 11%) was subtracted from the experimental values of  $\Delta\nu_{ij}^{\text{exp}}$  contained in the papers indicated above in compiling Table 1. The recomputation was carried out according to the formula

$$\Delta\nu_{ij}^{\text{H}_2\text{O}-x} (\text{cm}^{-1} \cdot \text{atm}^{-1}) = \left( \Delta\nu_{ij}^{\text{exp}} - \Delta\nu_{ij}^{\text{H}_2\text{O}-\text{H}_2\text{O}} \frac{P_{\text{H}_2\text{O}}}{760} \right) \frac{760}{P_x}, \quad (7)$$

where  $P_x$  and  $P_{\text{H}_2\text{O}}$  are the partial pressures (in mm Hg) of the expanding gas  $x$  and the water vapor which correspond to the value  $\Delta\nu_{ij}^{\text{exp}}$ . The half-widths  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{H}_2\text{O}}$  were taken in accordance with the experimental data displayed in Table 1.

As is evident from Table 1, the experimental values of the half-widths obtained for the majority of the lines by various methods and with different equipment are on the whole in good agreement with one another. True, there are also greatly differing results, but they "drop out of" the general picture which is given by Table 1. Such evidently erroneous results include data from the spectroscopic investigation [35] of the  $\lambda_{ij} = 0.538 \text{ mm}$  line (excluding the data for  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{H}_2\text{O}}$ ) and the measurements [18] of the half-width  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{H}_2\text{O}}$  of the  $\lambda_{ij} = 1.35 \text{ cm}$  line.

The values of partial half-widths of the  $\lambda_{ij} = 0.108 \text{ mm}$  line obtained spectroscopically in [34] are also not free from significant systematic errors. Actually, as shown in [36], the products  $S_{ij}\Delta\nu_{ij}$  ( $S_{ij}$  is the strength of the line) which were found in [34] for this line and were used subsequently to determine the half-widths  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{N}_2}$  and  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{H}_2\text{O}}$  are overestimated by a factor of  $\sim 1.5$ . Since the value of  $S_{ij}$  obtained by the authors of [34] from independent measurements evidently does not contain significant systematic errors,‡ it follows that the indicated half-widths should likewise be considered overestimated.

†Experimental data on the width of the  $\lambda_{ij} = 1.35 \text{ cm}$  line are of special interest for estimating the accuracy of the computation performed in [11], since the results of the first measurements of it in [17] were used in [11] to determine the free parameters entering into the computation formulas.

‡The fact that, just as for other lines that were investigated experimentally by various authors, the measured quantity  $S_{ij}$  for the  $\lambda_{ij} = 0.108 \text{ mm}$  line and also the values of  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{dry air}}$  and  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{H}_2\text{O}}$  obtained from the data of [36] using this value are in good agreement with theoretical data (viz., Tables 1 and 2) supports what has been stated.

TABLE 1. Experimental and Theoretical Values of the Half-Widths of the Absorption Lines of the Rotational Spectrum of Water Vapor

$\lambda, \text{mm}$ quantum transition	Expanding gas	T, K	$\Delta\nu_{1/2}^{\text{exp}}, \text{cm}^{-1} \cdot \text{atm}^{-1}$	$\Delta\nu_{1/2}^{\text{theo}}, \text{cm}^{-1} \cdot \text{atm}^{-1}$	$\frac{\Delta\nu_{1/2}^{\text{exp}}}{\Delta\nu_{1/2}^{\text{theo}}}$	Equipment; method of measurement*
1	2	3	4	5	6	7
13,483 5-1-6-5	Dry air	318	0,087 ± 0,01	0,0870	1	Measurement of $\gamma(\lambda)$ from the dependence of the Q of the multimode resonator on the air humidity; (~760) A. Measurement of the dispersion using a differential refractometer with a two-mode resonator (~1) B. A two-resonator refractometer (~2,5) As in item A As in item B A microwave spectrometer with a "wobbled" frequency and a waveguide cell, L = 1 m; (~0.1) As in item A; (0.1-20) As in item B; (0.03-30) Measurement of $\gamma(\lambda)$ by the variable-humidity method along a route L ~ 1 km long. A backward-wave tube (BWT) source - Present measurements Measurement of $\gamma(\lambda)$ under field conditions by the method of variable distance. BW I source C. A microwave spectrometer with double modulation and a waveguide cell, L = 1,2 m; (~0.4) D. Measurement of the tangent of the loss angle of the gas mixture using an installation with a Fabry-Perot resonator (~800) E. Measurement of $\gamma(\lambda)$ using a microwave-spectrometer installation with a vacuum vessel L = 150 m; (1-760) F. Measurement of $\gamma(\lambda)$ using an installation with an arc harmonic oscillator and a vessel L = 6 m; (10-760)
	N <sub>2</sub>	300	0,104	0,1013	1,03	
	O <sub>2</sub>	300	0,103	~0,041	1,02	
	"	300	0,064	"	1,56	
	"	300	0,058	0,4777	1,41	
	H <sub>2</sub> O	~300	0,357 ± 0,007	"	0,75	
	"	300	0,456 ± 0,005	"	0,95	
	"	300	0,446 ± 0,009	0,0960	0,93	
	"	300	0,102 ± 0,005†	0,0991	1,06	
	Dry air	284	0,102 ± 0,006	0,0975	1,03	
1,635 2-2-3-2	"	293	0,105†	0,1075	1,08	
	"	300	0,095 ± 0,001	"	0,88	
	N <sub>2</sub>	300	0,112 ± 0,006	"	1,04	
	"	300	0,090	"	0,84	
	"	300	0,111 ± 0,005	0,1087	1,02	
	"	300	0,089 ± 0,0008	~0,043	1,60	
	O <sub>2</sub>	300	0,068 ± 0,005	"	1,58	
	"	300	0,067	"	1,56	
	"	300	0,483 ± 0,005	0,4937	0,98	
	H <sub>2</sub> O	300	0,56 ± 0,06	0,5041	1,13	
"	~263	0,55 ± 0,1	"	1,09		

TABLE 1. (Continued)

1	2	3	4	5	6	7
0,922 4 <sub>0</sub> - 5 <sub>-4</sub>	Dry air	282	0,098 ± 0,008 [29]	0,0966	1,01	From measurements of $\gamma$ in the peak of the line using the theoretical strength $S_{theo} = 8,128 \text{ cm} \cdot \text{g}^{-1}$ of the line ( $\sim 760$ )
0,788 3 <sub>1</sub> - 4 <sub>-3</sub>	H <sub>2</sub> O Dry air	293 ~283 300 286	0,097 ± 0,005 [29] 0,55 ± 0,05 [27] 0,60 ± 0,05 [30] 0,101 ± 0,004 [31]	0,0943 0,5162 0,5071 0,0977	1,03 1,06 1,18 1,03	Measurement of $\gamma(\lambda)$ by the variable-humidity method; L = 363 m, BWT source As in item F; ( $\leq 10$ ) Measurement of $\gamma(\lambda)$ using a multipath vessel with a BWT; (1-25) Measurement of $\gamma(\lambda)$ by the variable-humidity method, L = 73 m, BWT source
0,538 1 <sub>-1</sub> - 1 <sub>1</sub>	N <sub>2</sub> H <sub>2</sub> O Dry air	295 299 293 303 300	0,091 ± 0,007 [32] 0,090 ± 0,007 [27] 0,55 ± 0,06 [27] 0,104 ± 0,004 [31] 0,11 ± 0,01 †	0,0958 0,1064 0,5385 0,1104 0,11115	0,95 0,85 1,02 0,94 0,99	Measurement of $\gamma(\lambda)$ using an installation with a vessel, L = 4,8 m, Arc harmonic oscillator source As in item F; ( $\sim 760$ ) Present measurements G. From spectrometric measurements [36] of the product $S_{ij} \Delta \nu_{ij}$ using $S_{ij}$ from the present measurements; ( $\sim 760$ )
0,398 2 <sub>-2</sub> - 2 <sub>0</sub>	N <sub>2</sub> O <sub>2</sub> H <sub>2</sub> O Dry air	~300 300 ~300 ~300 300 300	0,05 ± 0,01 [35] 0,06 ± 0,02 [36] 0,03 ± 0,01 [36] 0,5 ± 0,2 [38] 0,43 ± 0,05 † 0,102 ± 0,004 [34]	0,1245 0,05 0,4889 0,1044	0,45 0,43 0,60 1,02 0,88 0,98	Absorption measurements using an echelle spectrometer As in item G; (5-17) Measurement of $\gamma(\lambda)$ by the humidity-variation method, L = 0,9 m, BWT source; ( $\sim 760$ ) As in item G for $S_{ij}$ from [33]; ( $\sim 760$ ) As in item G for $S_{ij}$ from [33]; ( $\sim 760$ ) As in item G for $S_{ij}$ from [33]; ( $\sim 760$ ) As in item G for $S_{ij}$ from [34]; ( $\sim 760$ ) H. From measurements of the integral absorption (the Ladenburg-Reiche formalism); (300-760)
0,212 5 <sub>-3</sub> - 5 <sub>-1</sub>	H <sub>2</sub> O Dry air H <sub>2</sub> O	300 300 300	0,096 ± 0,01 † 0,44 ± 0,06 † 0,091	0,4648 0,0947 0,5123	0,92 0,95 0,96 0,89	Measurement of $\gamma(\lambda)$ by the humidity-variation method, L = 0,9 m, BWT source; ( $\sim 760$ ) As in item G for $S_{ij}$ from [33]; ( $\sim 760$ ) As in item G for $S_{ij}$ from [33]; ( $\sim 760$ ) As in item G for $S_{ij}$ from [34]; ( $\sim 760$ )
0,108 1 <sub>1</sub> - 2 <sub>1</sub>	Dry air N <sub>2</sub>	~300 ~300	0,105 ± 0,025 † 0,182 ± 0,036 [34]	0,1021 0,1144	1,03 1,59	H. From measurements of the integral absorption (the Ladenburg-Reiche formalism); (300-760)
0,0587 3 <sub>0</sub> - 4 <sub>1</sub>	H <sub>2</sub> O N <sub>2</sub>	~300 300	0,80 ± 0,16 † 0,56 ± 0,13 †	0,5256 0,0972	1,52 1,06	As in item G for $S_{ij}$ from [34]; (0,5-17)
0,0531 4 <sub>0</sub> - 5 <sub>0</sub>	H <sub>2</sub> O N <sub>2</sub> H <sub>2</sub> O	~300 ~300 ~300 ~300	0,122 ± 0,024 [34] 0,79 ± 0,14 [34] 0,129 ± 0,026 [34] 0,59 ± 0,12 [34]	0,4753 0,0999 0,4761	1,51 1,29 1,24	As in item H As in item H As in item G for $S_{ij}$ from [34]; (5-13)

\*The parentheses contain the pressure of the broadening gas in mm Hg.

†Due to neglect of the background of the wings of the other lines,  $\Delta \nu_{ij}^{exp}$  is overestimated by approximately 5%.‡The error  $\Delta \nu_{ij}^{exp}$  is the result of mean-square summing of the random errors of the measurements of  $(S_{ij} \Delta \nu_{ij})$  and  $S_{ij}$ .

In [36] the magnitude of this error is related to the insufficiently correct method of taking account of the overlap of spectral lines which was used by the authors in [34] in handling the experimental material. In this connection, the problem arises of the reliability of the measurements in [34] with respect to the half-widths of the  $\lambda_{ij} = 58.7$  and  $53.1 \mu$  lines as well, since the procedure for determining them was the same as that used for the  $\lambda_{ij} = 108 \mu$  line.

Let us make still another remark which applies to microwave-spectrometer measurements [21, 22, 26, 27]. As is evident from Table 1, the difference between the values of  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{N}_2}$  obtained in [21, 22, 26, 27] for the  $\lambda_{ij} = 1.635$  mm line is substantially greater than the random measurement errors. This indicates that even in measurements of this kind it is necessary to improve experimental procedures in order to obtain that accuracy of which the equipment is capable. It is of interest to compare the data of [21, 22, 26, 27] with the results of our measurements and the measurements of [23, 25] of the half-width  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{dry air}}$ . For this purpose it is necessary to use the values found for  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{N}_2}$  and  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{O}_2}$  in [21, 22, 26, 27] to calculate the value of  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{dry air}}$ . Comparison shows that the values of  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{dry air}}$  obtained on the basis of the data given in [22, 27] coincide with the results of [23, 25] and with the results of the present work; however, the data of [21, 26] lead to values of  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{dry air}}$  which are 15 to 20% lower.

Let us now go over to the comparison between experiment and theory. In accordance with the revision which Benedict and Kaplan [12] made of their computed data in [11] for  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{N}_2}$  we took the latter as the theoretical values of  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{dry air}}$ , while we took values 1.12 times greater than the tabulated values of [11] as the theoretical values of  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{N}_2}$ .\* The values of  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{H}_2\text{O}}$  and  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{O}_2}$  were taken from the calculations of [12]. The data on the temperature dependence which we used in calculating the theoretical half-widths corresponding to the temperatures for which the values of  $\Delta\nu_{ij}^{\text{exp}}$  were obtained are likewise contained in [11, 12].

Based on the data of Table 1, it may be concluded that for the quantities  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{dry air}}$ ,  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{N}_2}$ , and  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{H}_2\text{O}}$  the measured values coincide with the theoretical values within the limits of the experimental scatter. Specifically, for the half-widths  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{dry air}}$  which are of greatest significance in calculations of the atmospheric absorption, experiment substantiates the data of [11] within at worst 6%. With respect to the substantial (by a factor of  $\sim 1.5$ ) excess of the experimental data above the calculated data for the half-widths  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{O}_2}$  it follows that, as was noted by the authors of [12], due to the uncertainty of the values of the quadrupole moments of  $\text{O}_2$  the results of the calculations of  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{O}_2}$  in [12] are only crude.†

It should be noted that for a more complete check of the computations performed in [11, 12] it is desirable to have available experimental data for the rotational transitions corresponding to a wider range of quantum numbers  $J$  and  $\tau$  (the results of Table 1 correspond to values  $J$ ,  $|\tau| \leq 6$ ). In this respect, the measurements [6] of the half-widths of the lines in the interval  $\lambda = 14.5-21 \mu$  which correspond to the values  $5 \leq J \leq 13$  could serve as a useful supplement to Table 1, but regrettably the accuracy of these measurements was low. The comparison between the measured widths of 19  $\text{H}_2\text{O}$  lines with the calculated values cited in [11, 12] which was performed in [6] yielded values for the ratio  $(\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{air}})_{\text{exp}} / (\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{air}})_{\text{theo}}$  which varied from line to line from 0.8 to 1.9, while for  $(\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{H}_2\text{O}})_{\text{exp}} / (\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{H}_2\text{O}})_{\text{theo}}$  the values

\*The computation in [11] was performed in the approximation based on dipole-quadrupole interaction; the additional consideration of quadrupole-quadrupole interaction [46] yields noticeable corrections only for very weak lines.

†Using the relationship  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{dry air}} = P_{\text{N}_2} \Delta\nu_{ij}^{\text{H}_2\text{O}-\text{N}_2} + P_{\text{O}_2} \Delta\nu_{ij}^{\text{H}_2\text{O}-\text{O}_2}$  and the data of Table 1, it is not difficult to find that, for example, for the  $\lambda_{ij} = 1.35$  cm and  $\lambda_{ij} = 1.635$  mm lines an increase in  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{O}_2}$  by a factor of 1.5 leads to a change of only 4.8% in  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{dry air}}$ . This explains the fact that, notwithstanding the presence of a substantial divergence between  $(\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{O}_2})_{\text{exp}}$  and  $(\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{O}_2})_{\text{theo}}$ , it turns out that for coincidence of the measured and theoretical values of  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{N}_2}$  (or of  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{dry air}}$ ) these values also turn out to be close for  $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{dry air}}$  ( $\Delta\nu_{ij}^{\text{H}_2\text{O}-\text{N}_2}$ ).



TABLE 2. Experimental and Theoretical Values of the Strengths of the Lines of the Rotational Spectrum of H<sub>2</sub>O Vapors

Quantum transition	$\lambda_{ij}$ , mm	T, K	$S_{ij}^{\text{exp}}$ , cm·g <sup>-1</sup>	$S_{ij}^{\text{theo}}$ , cm·g <sup>-1</sup>	$\frac{S_{ij}^{\text{exp}}}{S_{ij}^{\text{theo}}}$
5 <sub>-1</sub> -6 <sub>-5</sub>	13,483	318	$1,40 \cdot 10^{-2}$ [17]	$1,382 \cdot 10^{-2}$	1,01
		300	$(1,42 \pm 0,03) \cdot 10^{-2}$ [39]	$1,407 \cdot 10^{-2}$	1,01
		300	$1,44 \cdot 10^{-2}$ [40]		1,02
2 <sub>2</sub> -3 <sub>-2</sub>	1,635	300	$2,77 \pm 0,17^*$ [25]	2,515	1,10
		284	$2,96 \pm 0,22$ p. m. †	2,780	1,06
		302	$2,24 \pm 0,13$ [26]	2,481	0,90
		286	$28,2 \pm 1,4$ [31]	28,76	0,98
3 <sub>1</sub> -4 <sub>-3</sub>	0,788	295	$26,3 \pm 1,3$ [32]	27,47	0,96
		299	$26,4 \pm 1,3$ [27]	26,97	0,98
		303	$1720 \pm 70$ p.m.	1671	1,03
1 <sub>-1</sub> -1 <sub>1</sub>	0,538	300	$1070 \pm 40$ [33]	1135	0,94
2 <sub>-2</sub> -2 <sub>0</sub>	0,398	~300	$(2,94 \pm 0,59) \cdot 10^4$ [34]	$3,278 \cdot 10^4$	0,90
1 <sub>1</sub> -2 <sub>1</sub>	0,108	~300	$(6,3 \pm 1,3) \cdot 10^4$ [34]	$6,436 \cdot 10^4$ ‡	0,98
3 <sub>1</sub> -4 <sub>1</sub>	0,0587	~300	$(1,76 \pm 0,35) \cdot 10^4$ [34]	$1,540 \cdot 10^4$	1,14
4 <sub>0</sub> -5 <sub>0</sub>	0,0531	~300			

\*Due to the neglect of the background from the wings of the other lines during the determination of  $\gamma_{ij}^{\text{exp}}(\nu_{ij})$  and  $\Delta\nu_{ij}^{\text{exp}}$ , the value of  $S_{ij}^{\text{exp}}$  is overestimated. According to estimates, the correction amounts to about 10%.

†The letters p.m. denote the results of the present measurements.

‡In [34] the theoretical value of  $S_{ij}$  is indicated to equal  $5.191 \cdot 10^4$  cm·g<sup>-1</sup> (38.01 cm<sup>-2</sup>·atm<sup>-1</sup>) for some ununderstood reason.

varied from 0.6 to 1.7. However, notwithstanding such large deviations of the measured half-widths from the calculated values, the authors of [6], having in mind the insufficient accuracy of the results which they obtained, did not find it possible to draw any conclusion concerning the existence of the discrepancy between experiment and computation. It may also be added that if one speaks of the average (over all the lines) deviations of the experimental data [6] from the theoretical results, then it amounts to ~25% for the half-widths  $\Delta\nu_{ij}^{\text{H}_2\text{O-air}}$  and is close to zero for  $\Delta\nu_{ij}^{\text{H}_2\text{O-H}_2\text{O}}$ .

The Strength of the Lines. The strength (integral intensity) of a line is by definition

$$S_{ij} = \int_0^{\infty} \gamma_{ij}(\nu) d\nu. \quad (8)$$

In view of: a) the unbounded nature of the domain of integration; and b) the overlap of the spectral lines, it is difficult to perform direct measurements of the quantity  $S_{ij}$ . Therefore, indirect methods of determining it are used.

In those cases when the resolution of the equipment is sufficient to perform direct measurements of: 1) the absorption coefficient in the peak of the  $\gamma_{ij}(\nu_{ij})$  line; and 2) the half-width  $\Delta\nu_{ij}$  of the line (for example, in the present measurements), the value of  $S_{ij}$  may be found from the relationship

$$S_{ij} = \pi \gamma_{ij}(\nu_{ij}) \Delta\nu_{ij}. \quad (9)$$

It derives from (8) when the spectral line has the form (5).

By now, such a method has been used to determine the strength of the following lines:  $\lambda_{ij} = 13.483$ ; 1.635; 0.788; 0.538; and 0.398 mm. Their values are displayed in Table 2.

For the shortwave lines in Table 2, which were studied by the infrared spectroscopy method in [34], the values of  $S_{ij}^{\text{exp}}$  were determined by another method based on measuring the integral absorption in the so-called linear region  $[(1/2)\gamma_{ij}(\nu_{ij})\rho L \ll 1]$  and using the known Ladenburg-Reiche [47] expression.

We performed the calculation of the theoretical values of  $S_{ij}$  indicated in Table 2 using the formulas derived on the basis of (9) by means of Eq. (6) of [14]:

$$S_{ij} \text{ (cm} \cdot \text{g}^{-1}\text{)} = \frac{1,393 \cdot 10^{40} \mu^2 \beta_{ij}}{G(T) \lambda_{ij}} \left| \exp\left(-\nu_i \frac{hc}{kT}\right) - \exp\left(-\nu_j \frac{hc}{kT}\right) \right|, \quad (10)$$

where  $\mu$  is the dipole moment of the H<sub>2</sub>O molecule;  $\beta_{ij}$  is the oscillator force of the quantum transition;  $G(T)$  is the statistical sum over the rotational states;  $\nu_i$  and  $\nu_j$  are the energy terms of the i-th and j-th

states. For the quantity  $\mu$  we adopted the value  $1.8546 \cdot 10^{-18}$  CGSE in accordance with the latest measurements [48, 49] of Stark splitting. The values of  $\beta_{ij}$  were determined on the basis of tabulated data [14];  $G(T)$  was calculated according to the approximation formula [50]

$$G(T) = 3.4264 \cdot 10^{-2} T^{3/2}, \quad (11)$$

while the energy terms were taken from [8].

A comparison of the data in Table 2 shows that the theoretical calculations of the strengths of the line for small quantum numbers  $J$  are well substantiated by experiments. Specifically, for lines studied by radio methods ( $\lambda_{ij} \geq 0.398$  mm) the difference between the average measured (for each line) values of  $S_{ij}$  and the calculated values, just as for the half-widths, amounts to  $\lesssim 6\%$ .

As far as an experimental check of the calculations of the strength of the line corresponding to large values of  $J$  is concerned, here we may merely refer to the measurements in [6] in the infrared range which were mentioned above. In accordance with [6], the ratio between the measured values of  $S_{ij}$  and the values calculated on the basis of the rigid-asymmetrical-top model for the investigated 19 lines lies in the 0.8 to 1.5 range. The authors of [6] also calculated the strengths of these lines with allowance for centrifugal perturbations, but its agreement with experiment did not undergo an overall improvement in comparison with the preceding case (here  $S_{ij}^{\text{exp}}/S_{ij}^{\text{theo}} = 0.95-1.7$ ). Evidently, the difference between the measured and calculated values of  $S_{ij}$  is caused here not so much by an inaccuracy of the theoretical calculations as by experimental errors.

In conclusion, the authors thank V. I. Anikin, B. A. Sverdlov, and A. I. Khvostova for their help in the preparation and performance of the measurements, and S. A. Zhevakin for discussing the results and going over the manuscript.

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