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LASER SPECTROSCOPY

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Determination of spectral width of laser lines in the IR range using the absorption spectroscopy method

O.K. Voitsekhovskaya, D.V. Volkov, D.E. Kashirskii, V.S. Korchikov

Abstract. **An engineering technique for determining the spectral width of laser lines is proposed, based on the attenuation of the radiation in a cuvette with known gas composition under fixed thermodynamic conditions using absorption spectroscopy. Preliminarily, based on the theoretical analysis and computational procedures, the dependence of the gas transmission function on the laser linewidth is specified for a particular laser transition, and then, using the nomographic chart and the experimental value of the function, the linewidth of the laser line is found. In the remote gas analysis using the differential absorption method the correction of the measured value of the probed gas absorption coefficient is carried out, taking the spectral width of the laser radiation linewidth into account. The technique requires provision with appropriate spectroscopic information. Particular consideration is carried out for the radiation of a carbon monoxide laser, absorbed by water vapour**

Keywords: *radiation, absorption, laser linewidth, CO laser, water vapour.*

1. Introduction

The absence of laser radiation linewidth control may cause significant errors in measuring the spectral characteristics of gaseous media by means of the remote probing method. In many cases just this is the reason, why the absorption coefficients of gases measured at the same laser line appear to be different [1].

The spectral width of the laser radiation line depends on many factors. The variations of the linewidth arise from the difference in the laser oscillation conditions and may be conventionally divided into two groups, the physical and the technological ones. The first group is determined by the line-broadening effects in the active medium of the laser, caused by the collision processes and the Doppler effect, while the second group is due to the instrument effects, namely, the cavity properties and other factors, e.g., mirror vibration, etc. The line profile shape of the radiation output from a laser is often indefinite, because the high degree of monochromaticity does not allow high-precision detailing of its spectrum.

The existing methods used to determine the spectral width of laser lines are complicated and their implementation requires high financial expenditure. They include interferometric methods, heterodyning methods, and methods based on measuring the statistical characteristics of laser noise. However, these methods are research-oriented and require the use of additional devices, whereas the methods of measuring the composition and concentration of gases, e.g., in the Earth atmosphere or in other gas volumes, using differential absorption approach, may be considered as routine ones.

In the present paper we describe a simple method of laser linewidth determination, applicable even under the field conditions. It is proposed to used cuvettes with calibrated volume of gas (with fixed concentration and temperature) to determine the spectral width of a laser line by measuring the transmission function (TF) of the laser radiation by using a preliminarily calculated theoretical dependence of TF on the spectral width of the radiation line. In the literature the method, based on the TF measurement in a gas medium under the modulation of its optical thickness or its pressure, is applied for experimental determination of the spectral shape of the radiation line of the tunable laser trass absorption gas analyser in the three-micrometre range ($\lambda = 2.5-4.4$ μm) [2]. An analogous proposal on using the gas cuvettes with known thermodynamic parameters, but for measuring the difference of wavelengths of radiation from Nd:YAG laser after passing through the nonlinear crystal $LiNbO_3$ was made in [3]. Methane and formaldehyde were used as reference gases. The frequency dependences of the TF of these gases in a cuvette with dry air were theoretically calculated with high resolution, and from the measured absorption in the cuvette the wavelength of the source radiation was determined.

The effect of the laser linewidth on the TF can be described by the relation

$$\tau = \frac{\int \mathrm{d}\nu A(\nu,\Delta\nu)\exp[-f(\theta,p,\nu,L)]}{\int \mathrm{d}\nu A(\nu,\Delta\nu)}, \tag{1}$$

where ν is the wave number; $\Delta\nu$ is the spectral width of the laser line; $A(\nu,\Delta\nu)$ describes the profile of the laser oscillation line; $f(\theta,p,\nu,L)$ is the spectral optical thickness of the gas in the cuvette; θ and p are the temperature and the pressure of the absorbing gas in the cuvette; L is the length of the cuvette.

The particular value of the linewidth of the radiation line is rarely discussed in the papers on the interaction of laser radiation with matter. However, for correct processing of experimental results it is necessary first of all to determine the spectral width of the laser line. The influence of the width of the laser line on the measured absorption coefficient and the importance of determining this characteristic in the gas analysis problem are illustrated in Fig. 1.

O.K. Voitsekhovskaya, D.V. Volkov, D.E. Kashirskii, V.S. Korchikov
Tomsk State University, prosp. Lenina 36, 634050 Tomsk, Russia;
e-mail: vok@elefot.tsu.ru

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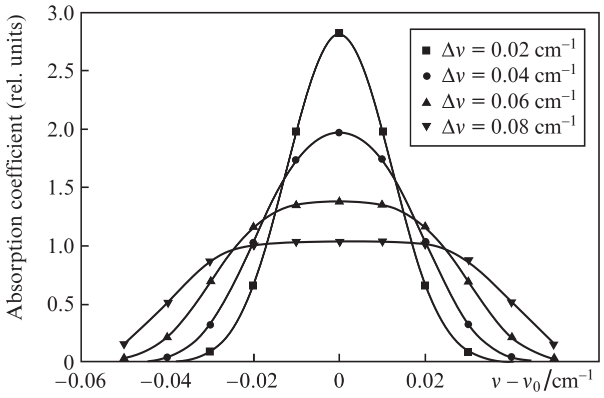

Figure 1. The effect of laser linewidth $\Delta\nu$ on the measured absorption coefficient for coincident centres (ν_0) of the laser radiation and the water vapour absorption lines. The spectral half-width of the absorption line centred at the frequency 1901.76001 cm^{-1} is equal to 0.039 cm^{-1} at the temperature 1000 K (see Table 1).

2. Determination of the CO-laser radiation linewidth by measuring its absorption in water vapour

The proposed approach is considered by the particular example of estimating the influence of CO laser radiation linewidth on the magnitude of its absorption by water vapour. Let us describe the implemented scheme of plotting the dependence of TF on the width of the laser line for CO laser. Having two data arrays (the frequencies of line centres of laser radiation and the parameters of spectral lines of water vapour), one can easily choose vibrational–rotational (VR) transitions of water vapour, the frequencies of which are close to those of laser lines. Using the developed computational schemes for informational support of modelling the remote monitoring of thermodynamically nonuniform gas media [4], the transmission of H_2O is calculated for different laser linewidths at fixed temperature and gas concentration. The shape of the laser radiation line is described by a Gaussian profile, and the absorption line is taken to have a Voight profile.

The line centres of laser radiation should be determined with sufficient accuracy, no worse than 0.0001 cm^{-1}. However, while the energies of the VR levels for the vibrational states with $v \leqslant 3$ are known with the necessary precision, the evaluation of energies of higher levels with $v \geqslant 7$, which correspond to the transitions close to the lines of CO laser radiation, requires additional studies. Many publications are devoted to this issue. For example, in [5] a thorough analysis of the known data on the VR energy levels is performed and an array of the energy values of VR levels with $v \leqslant 41$ and $J \leqslant 60$ is proposed.

Earlier the authors of the present paper determined the Dunham coefficients for the ground electronic state of CO molecule, based on the newly obtained values of the effective rotational and centrifugal constants, and predicted the energies of high VR states [6]. A special feature of our approach is the use of the energies of VR levels, calculated by means of the traditional polynomial formula, as the initial data for the determination of Dunham coefficients. Following the scheme, analogous to that of Ref. 6, we recalculated the Dunham coefficients using the values of the effective rotational and centrifugal constants of the CO molecule [7] and, correspondingly, the positions of centres of the laser oscillation lines.

The calculation of absorption coefficients of the reference gas (in our example water vapour) and the precision of determination of the spectral line parameters are worth a separate consideration. In the calculations of the absorption coefficient in the case of a uniform medium (the temperature and gas concentration being constant along the path) the key parameters are those of the absorption lines of the gases, since the problem of line profile choice is solved by using the Voight profile. The known databases on the spectral line parameters (SLP) for the spectroscopy of gaseous media, elaborated by a number of leading research centres of the USA [HITRAN (http://www.hitran.com), JPL (http://spec.jpl.nasa.gov), SpectralCalc (http://www.spectralcalc.com/spectralcalc.php)], France [GEISA (http://ara.lmd.polytechnique.fr)], and Great Britain (http://www.ucl.ac.uk) contain the data only for normal conditions (T = 296 K, p = 1 atm). The access to the database HITEMP (http://www.cfa.harvard.edu/hitran/HITEMP.html) [8], containing the parameters of spectral lines of five gases (CO, CO_2, H_2O, NO, OH) for high-energy VR states, has shown that this base not only requires development of original software to calculate the spectral characteristics, but also needs full testing and data verification.

Below we present the results of calculation of transmission function of water vapour depending on the linewidth of the CO laser radiation for two laser lines. In Figs 2 and 3 we

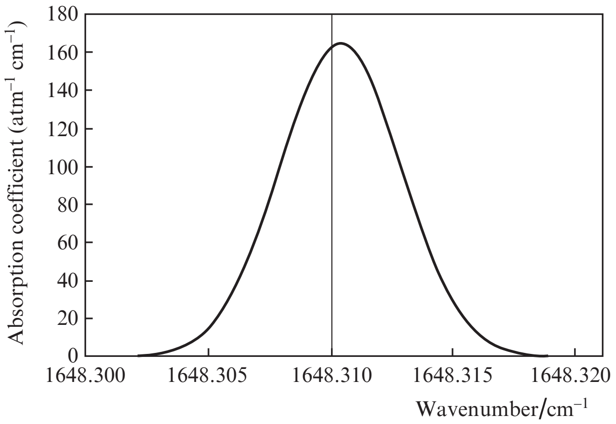

Figure 2. Spectral dependence of the absorption coefficient of H_2O at the VR transition of the ν_2 band ($J' = 2, K'_A = 2, K'_C = 0 \leftarrow J'' = 2, K''_A = 1, K''_C = 1$) centred at the frequency 1648.3104 cm^{-1} at the pressure 0.001 atm, temperature 296 K and cuvette length 5 cm. Vertical line shows the centre of the CO laser oscillation line ($v' = 17, J' = 22 \rightarrow v'' = 16, J'' = 21$) at the frequency 1648.310 cm^{-1}.

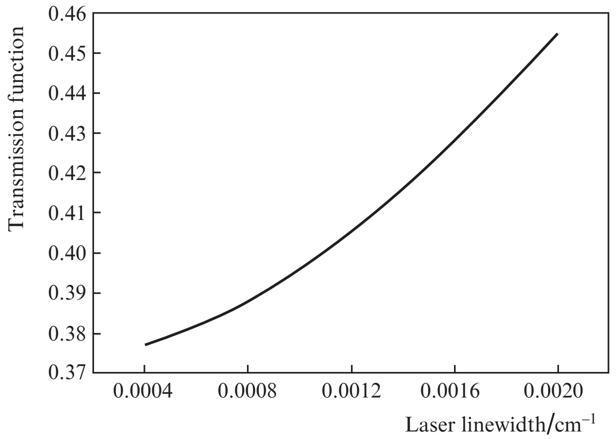

Figure 3. Dependence of TF on the spectral width of laser radiation line under the conditions of Fig. 2.

show the results of calculations for the line of CO laser radiation and the absorption line of water vapour with practically coincident centres, namely, the centre of the absorption line of H_2O at the VR transition of the ν_2 band ($J' = 2$, $K'_A = 2$, $K'_C = 0 \leftarrow J'' = 2$, $K''_A = 1$, $K''_C = 1$) is equal to 1648.3104 cm^{-1}, while the frequency of the centre of the CO-laser oscillation line ($v' = 17$, $J' = 22 \rightarrow v'' = 16$, $J'' = 21$) is equal to 1648.310 cm^{-1}. In this case the dependence of the Gaussian-shaped laser linewidth on the TF in the 5 cm-long cuvette is approximated by the polynomial

$$y = -0.21135 + 1.446x - 3.30398x^2 + 2.54189x^3, \qquad (2)$$

where x is the transmission function, y is the laser linewidth (in cm^{-1}).

Thus, when the laser line centre coincides with the centre of the absorption line of the reference gas, an increase in the probed gas transmission with the growth of the laser linewidth is observed (Figs 2 and 3). This dependence corresponds to Fig. 1: The radiation spectrum becomes broader, and the transmission of the gas medium increases due to the small absorption at the wings of the absorption line.

In the case, when the centre of the radiation line coincides with the edge of the absorption line, the growth of the laser line spectral width causes a decrease in the reference gas transmission (Figs 4 and 5), i.e., the line broadening leads to 'covering' of the absorption line by the radiation spectrum. In this case the polynomial for calculating the laser linewidth takes the form

$$y = 0.1301 - 0.46477x + 0.56854x^2 - 0.2362x^3. \qquad (3)$$

The maximal error of the approximations (2) and (3) is 3.5%.

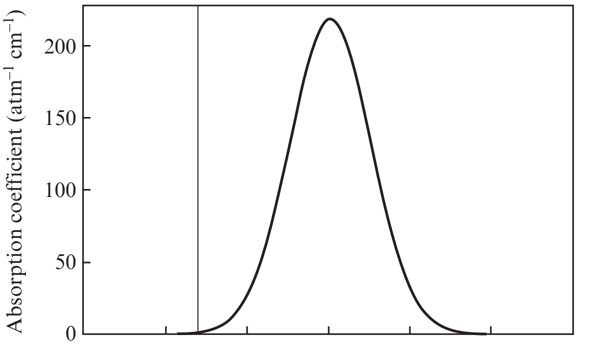

Figure 4. Spectral dependence of absorption coefficient of H_2O at the VR transition of the ν_2 band ($J' = 5$, $K'_A = 1$, $K'_C = 4 \leftarrow J'' = 5$, $K''_A = 0$, $K''_C = 5$) centred at the frequency 1675.5151 cm^{-1} at the pressure 0.1 atm, temperature 296 K and cuvette length 10 cm. Vertical line shows the centre of the CO laser oscillation line ($v' = 17$, $J' = 14 \rightarrow v'' = 16$, $J'' = 15$) at the frequency 1675.507 cm^{-1}.

3. Interpretation of the results of measuring the absorption of the CO laser radiation

Below we consider a particular example of interpretation of the experimental results taken from Ref. [9], in which the unique measurements of the absorption of radiation by gas are presented. The measurements were performed at a single CO laser line ($v' = 7$, $J' = 14 \rightarrow v'' = 8$, $J'' = 15$) under different thermodynamic conditions of the absorbing medium, the water vapour at high temperatures (1300–2300 K) and different pressures (0.28–1.22 atm). Our goal is to demonstrate that the radiation linewidth affects the transmission through the medium, and to substantiate the necessity of preliminary analysis of the available spectroscopic information for the calculation of the TF dependence on the laser linewidth.

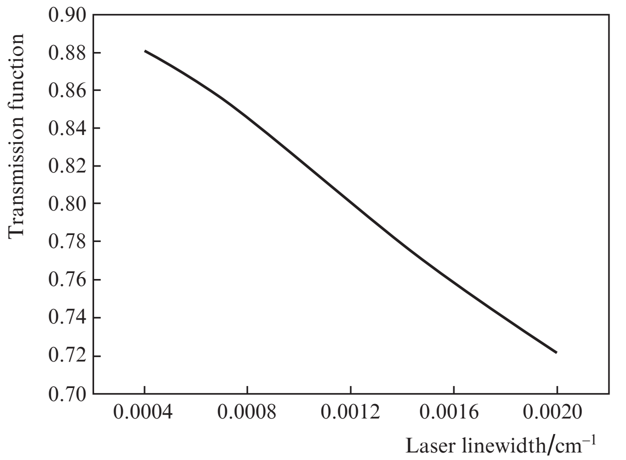

Figure 5. Dependence of the water vapour TF on the laser radiation spectral linewidth under the conditions of Fig. 4.

As already mentioned, we carried out the redetermination of the spectroscopic constants for calculating the energies of VR levels of carbon monoxide. That is why the centre frequency of the radiation line for the transition $v' = 7$, $J' = 14 \rightarrow v'' = 8$, $J'' = 15$ is accepted to be 1901.7570 cm^{-1}, in contrast to the value 1901.762 cm^{-1}, used in [9]. Using the results of Ref. [5], in which the energies of the VR levels of the CO molecule are presented, for the centre frequency of this transition the value of 1901.7616 cm^{-1} was obtained. We emphasise again, that it is desirable to have the precision of determination of the laser line centre frequency not worse than 0.0001 cm^{-1}. In the future we plan to prepare a set of recommended centre frequencies of the CO laser radiation lines after comparing the results of calculation of the absorption coefficients for all oscillation wavelengths, choosing the results of experiments on measuring the absorption of CO laser radiation by different gases as a criterion.

As to the possible values of the laser linewidths, according to [10], in practically implementable working regimes of the CO laser operation the collision broadening of laser transition lines is smaller than 20 MHz (0.00067 cm^{-1}) and much smaller than the Doppler linewidth, which at temperatures from 77 to 300 K lies within the interval from 70 to 140 MHz (0.0023–0.0047 cm^{-1}). These values correspond to Doppler linewidths, calculated following the formula

$$\Delta\nu_D = 4.3\times10^{-7}\nu_0(T/m)^{1/2}, \qquad (4)$$

where ν_0 is the linewidth of laser radiation line (in cm^{-1}); T is the temperature (in K); m is the mass of the molecule (in a.m.u.).

The unique information retrieval database 'Gas radiation' [4] served as a source of spectroscopic information on SLP of water vapour and the programme code for our TF calculations. Its major advantage is the possibility to create databases on SLP of gaseous combustion products (CO, NO, H_2O, CO_2) in the IR range within the temperature interval from 300

Table 1. Parameters of the VR absorption lines of water vapour at the pressure 1 atm, taken from the databases HITEMP-2010 (italic) and Gas radiation.

T/K	Line centre frequency/cm^{-1})	Intensity/ cm^{-2} atm^{-1}	Line half-width/cm^{-1}	Lower level energy/cm^{-1}	Transition $v_1'v_2'v_3' \leftarrow v_1''v_2''v_3''; J'K_A'K_C' \leftarrow J''K_A''K_C''$
1000	1901.56055 *1902.33663*	0.00019 *0.00041*	0.025 *0.035*	4428.11 *4428.114*	010 ← 000; 18 06 13 ← 17 07 10
	1901.61670 *1901.54773*	9.72×10^{-5} 4.55×10^{-5}	0.020 *0.031*	4716.31 *4716.379*	011 ← 020; 10 02 09 ← 11 02 10
	1901.68896 *1901.68896*	0.00189 *0.000415*	0.044 *0.032*	5286.56 *5286.563*	040 ← 030; 06 04 03 ← 05 03 02
	1901.69434 *1902.21060*	0.00026 *0.00037*	0.025 *0.023*	3824.5 *3824.49*	010 ← 000; 16 07 09 ← 15 08 08
	1901.76001 *1901.75951*	0.12 *0.1*	0.018 *0.039*	1690.67 *1690.66*	010 ← 000; 12 03 10 ← 11 02 09
2000	1901.56055 *1902.33663*	0.00033 *0.00091*	0.018 *0.032*	4428.11 *4428.114*	010 ← 000; 18 06 13 ← 17 07 10
	1901.61670 *1901.54773*	0.000269 *0.000124*	0.014 *0.034*	4716.31 *4716.379*	011 ← 020; 10 02 09 ← 11 02 10
	1901.62402	3.94×10^{-5}	0.016	7966.68	012 ← 021; 08 02 07 ← 09 02 08
	1901.68896 *1901.68896*	0.0079 *0.0017*	0.031 *0.019*	5286.56 *5286.563*	040 ← 030; 06 04 03 ← 05 03 02
	1901.69434 *1902.21060*	0.00028 *0.00053*	0.018 *0.023*	3824.5 *3824.49*	010 ← 000; 16 07 09 ← 15 08 08
	1901.75391	1.64×10^{-5}	0.033	9394.98	003 ← 012; 04 01 03 ← 05 01 04
	1901.76001 *1901.75951*	0.029 *0.030*	0.012 *0.038*	1690.67 *1690.66*	010 ← 000; 12 03 10 ← 11 02 09
	1901.82520	4.86×10^{-6}	0.033	9411.63	003 ← 012; 04 02 03 ← 05 02 04

to 3000 K. This allows direct calculation (using the line-by-line method) of the spectral characteristics of gases, providing the results with maximal precision.

The analysis of SLP of water vapour revealed significant disagreement of their values taken from the databases 'Gas radiation' and HITEMP-2010. These values are summarised in Table 1 for the frequency interval from 1901.5 to 1902.5 cm^{-1} (the water spectral lines that provide major contribution to the absorption at the given laser line and have the intensities not less than 10^{-5} cm^{-2} atm^{-1}). From Table 1 it is seen that the intensities and half-widths of the lines, as well as the number of lines, taken from the databases [8] and [11], are different.

The closest to the centre of the CO laser radiation line, studied in [9], is the VR absorption line of H_2O with the centre at the frequency 1901.76001 cm^{-1} ($v_1' = 0$, $v_2' = 1$, $v_3' = 0$, $J' = 12$, $K_A' = 3$, $K_C' = 10 \leftarrow v_1'' = 0$, $v_2'' = 0$, $v_3'' = 0$, $J'' = 11$, $K_A'' = 11$, $K_C'' = 2$). The authors of Ref. [9] proposed for this line the following temperature dependence of the half-width:

$$2\gamma = 0.027\left(\frac{1300}{T}\right)^{0.9}. \qquad (5)$$

At the same time, for the half-width caused by collisional processes the dependence on the temperature and the pressure of the absorbing medium is described by the relation

$$\gamma = \gamma_0 \frac{p}{p_0}\left(\frac{T_0}{T}\right)^n, \qquad (6)$$

where the exponent n is most frequently taken to be 0.5, which was used in our calculations.

The analysis of the modern databases HITEMP-2010 and HITRAN-2008 [8] for the considered H_2O line yields a different value of the exponent in the temperature dependence of the half-width, namely, $n = 0.05$. In Ref. [12] for this line $n = 0.372$ is obtained, but it should be noted that the temperature interval, considered in this work, is 240–388 K.

Taking the above considerations into account, we performed the calculation of the optical thickness of the water vapour for the CO laser radiation line centred at the frequency 1901.758 cm^{-1} ($v' = 7$, $J' = 14 \rightarrow v'' = 8$, $J'' = 15$) under the conditions of the experiment [9]. The agreement of the results of our calculations with the experimental values of the optical thickness at real values of the laser radiation linewidth within the error of the experiment confirms that the TF at the stable laser wavelength depends on the laser radiation linewidth. Table 2 summarises the CO laser radiation linewidths, at which the calculated optical thickness demonstrates the

Table 2. Probable values of the CO laser radiation linewidth for the measured values of the optical thickness of the water vapour at different thermodynamic conditions and the cuvette length 15.24 cm.

Temperature/ K	Pressure/ atm	Laser radiation linewidth/ cm^{-1}	Optical thickness		
			Calculation	Experiment [9]	Δ (%)
1327	0.64	0.0034	26.25	24.27	8.2
1335	0.64	0.0034	25.80	24.55	5.1
1590	0.28	0.0040	12.46	11.52	8.2
1603	0.55	0.0034	17.49	17.59	0.6
1752	0.66	0.0035	15.03	15.09	0.4
1840	0.84	0.0045	13.70	15.97	14.2
1840	0.80	0.0048	13.80	14.82	6.9
1855	1.05	0.0040	14.24	16.91	15.8
1924	0.78	0.0040	12.20	13.36	8.7
1930	0.65	0.0040	11.90	12.28	3.1
1959	1.02	0.0035	12.03	13.72	12.3
2003	0.80	0.0048	10.63	12.32	13.7
2064	0.76	0.0020	9.83	11.43	14.0
2094	0.77	0.0036	9.28	10.38	10.6
2199	0.76	0.0036	8.28	10.25	19.2
2316	0.69	0.0036	6.82	7.92	13.9

Note: Δ is the difference of the calculated values from the experimental data.

best agreement with that obtained in the experiment [9]. The laser radiation linewidth varied within the limits from 0.0020 to 0.0044 cm^{-1}, which corresponds to the Doppler linewidths at the temperatures 77–300 K.

A clear example of the effect of the laser linewidth on the absorption coefficient at different temperatures is presented in Fig. 6. It is seen that the dependences of the absorption coefficient on the temperature possess similar form for different laser linewidths, but with the growth of temperature the discrepancy between the absorption coefficient values decreases. The experimental value of the absorption coefficient is presented in Fig. 6 for verification of calculations.

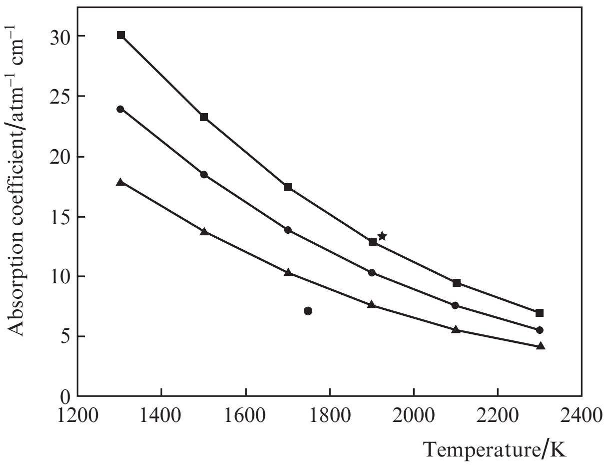

Figure 6. Temperature dependences of the water vapour absorption coefficient for CO laser radiation line centred at the frequency 1901.758 cm^{-1} ($v' = 7$, $J' = 14 \rightarrow v'' = 8$, $J'' = 15$), the linewidth being equal to 0.02 (▲), 0.002 (●), 0.04 cm^{-1} (■), and the experimental value of the water vapour absorption coefficient at the temperature $T = 1924$ K and the pressure $p = 0.78$ atm [9] (★).

The use of high-temperature gases in the discussed problem is not recommended because of the difficulty of practical implementation, especially as the form of the dependence of the TF on the spectral width for the same laser line is kept the same under normal temperatures too, which is reflected in Fig. 7.

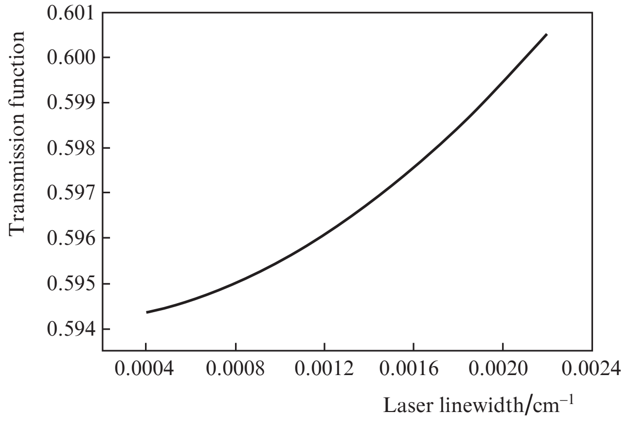

Figure 7. Dependence of the water vapour TF on the CO laser oscillation linewidth centred at the frequency 1901.758 cm^{-1} ($v' = 7$, $J' = 14 \rightarrow v'' = 8$, $J'' = 15$) at normal temperature (296 K).

The precision of determining the laser linewidth by means of the described technique depends mainly on the characteristics of the modern photodetectors. The smaller is the measurement error for the CO laser radiation power (achieving 100 kW [13–15] at the cavity exit) after passing through the cuvette, the higher is the precision of the laser linewidth determination. For example, the pyroelectric sensor manufactured by the company OPHIR has the power measurement error ~2% [16].

4. Recommendations on the choice of gas components for the lines of other lasers

Generally, it is possible to formulate the criteria of choosing the test gases and their reference absorption lines for determination of the laser radiation linewidths. First, the laser radiation line centre frequency should be known with the accuracy of at least 0.0001 cm^{-1} in the mid-infrared region, and the radiation frequency should be kept stable. Otherwise the change of the frequency separation between the centres of the absorption line and the laser line will lead to the change of TF. Second, the thermodynamic conditions in the cuvette should be stable, since the parameters of the polynomial dependence are found for definite temperature and concentration of the reference gas. For the deviation of temperature by 0.1 K the error of the reconstructed laser linewidth using the polynomial dependences (2) and (3) amounted, in average, to 1.5% and 0.7%, respectively. The change of the pressure in the cuvette by 1% leads to the mean error of reconstruction 1% for the dependence (2) and 1.2% for the dependence (3).

The proposed method also implies high requirements to the accuracy of determining the SLP of the absorbing gases. Moreover, the reference gas should have relatively simple spectrum with minimal overlap of lines and, in principle, be easy in experiment, i.e., have low cost and not interact with the cuvette material. The advantages of using the water vapour in the present work consist in the fact that its spectrum in the IR region is thoroughly studied, and the centre frequencies of the absorption lines are determined with high precision (the method of Fourier spectroscopy allows measurement of the centre frequencies of absorption lines with the error less than 10^{-6} cm^{-1}).

The existing methods of radiation detection impose limitations on the use of the proposed technique. The necessity of complex account of the characteristics of the triad 'source–medium–detector', when choosing the most efficient algorithm for diagnostics of the laser radiation linewidth, makes one to vary thermodynamic conditions in the medium in order to provide the maximal precision of linewidth determination. Moreover, the use of the described scheme of plotting the dependence of the TF on the laser linewidth requires appropriate spectroscopic information software, developed by one of the authors during many years (see, e.g., [11, 17, 18]).

In conclusion, we recall again that earlier [1] the results of the analysis of the experimental data on the absorption coefficients for the CO_2 laser radiation in water vapour and ammonia were reported and the dependences of the absorption coefficients for NH_3 and H_2O on the laser radiation linewidth were presented. These data may be used to determine the carbon dioxide laser radiation linewidth after adjusting the spectroscopic information on SLP of the considered gases.

Unavoidable errors of measuring the pressure, temperature, optical path length, and signal-to-noise ratio reduce the accuracy of the laser radiation linewidth. However, being aware of the difficulties of measuring the laser line parameters [19], we can recommend the proposed technique as an alternative method of determination of laser linewidths.

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