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Determination of the optimal set of absorption lines for detection of the maximum temperature in a spatially non-uniform media by absorption spectroscopy with diode lasers

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Determination of the maximum temperature in spatially non-uniform media by absorption spectroscopy with diode lasers is nontrivial problem. When thermodynamic equilibrium (TDE) is fulfilled the temperature of a uniform media is defined by fitting the experimental line profiles with the ones simulated basing on the spectroscopic data bases. In this simple case, two frequency-tunable diode lasers operating in the range of two strong lines with different energies of the lower levels are sufficient. However, in an inhomogeneous environment, the shape of the absorption line of the test molecule is resultant along the entire sensing path, which increases the number of parameters necessary to characterize the object. This leads to an increase in the number of detected lines (the number of diode lasers), an increase in the cost of the system, and a complication of the algorithm for processing experimental spectra. The article considers the case, which is important for the diagnosis of hot media, when it is necessary to determine not the exact profile of the temperature distribution, but only its maximum value. A minimum set of absorption lines has been determined, which can be used to estimate the maximum temperature under the assumption of its trapezoidal distribution in the medium along the sensing line. Combinations of four lines were found to solve the problem, and the accuracy of the maximum temperature estimate was evaluated. To determine the integral intensity of absorption lines in an inhomogeneous medium, a method is proposed for fitting the shape of the line with two Voigt profiles. It is shown that in the case of recording the minimum temperature at the boundary of the hot zone using commercial thermocouples, it is possible to use only three absorption lines. In this case, however, the error in determining the maximum temperature increases by about 4 times compared to the registrations of the four lines.

Keywords: absorption spectroscopy, diagnostic of hot zones, non-uniform spatial distribution, fitting of absorption lines profiles.

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Introduction

The method of diode laser absorption spectroscopy (DLAS) is one of the most widely used techniques in laser spectroscopy for diagnosing gas media [1–4]. The key advantage of laser methods is the non-perturbing mode of obtaining information about the parameters of the probed medium. For example, when determining the temperature of hot gas objects, especially hot gas flows, traditional thermocouples cannot be used, as introducing any physical object into the probed region substantially alters the thermal fields and flow structures near the inserted body, thereby distorting the measurement results. In contrast, the beam of a low-intensity diode laser (DL) does not affect the properties of the probed medium at all, ensuring the accuracy of the diagnostic results.

A second important advantage of methods using DLs is the ability to diagnose remote objects or „dangerous“ zones for personnel, such as combustion zones or nuclear reaction zones. The DL radiation can either be shaped by optical systems into a low-divergence beam and delivered to the object, or transmitted to the probed object via optical fibers over sufficiently long distances.

Determination of the medium temperature by the DLAS method is based on measuring the integrated absorption on several lines of a test molecule with different lower-level transition energies. Under thermodynamic equilibrium (TE) in the medium, the ratios of the integrated intensities of the test molecule's absorption lines obey the Boltzmann law and are determined by the medium temperature. At gas pressures above ~ 0.1 atm TE is established on a timescale of order 10^{-4-5} s. The characteristic timescales for changes in density and temperature in high-speed gas flows of modern aircraft engines ($> 10^{-4}$ s) allow the use of the TE model in the medium, and the DLAS method enables accurate determination of the temperature dynamics with a temporal resolution of no worse than $\sim 10^{-3}$ s.

Two variants of the DLAS method have been developed. The first is based on measuring the direct absorption of the probing radiation. This method performs well at high signal-to-noise ratios. At high noise levels, various variants of modulation spectroscopy work better, in which the frequency of the probing diode laser radiation is modulated, and absorption is recorded at the harmonics of the modulation frequency. The capabilities of the DLAS method, both in direct absorption mode and using various modulation spectroscopy variants, have been successfully demonstrated in numerous studies and reviews [2–11].

Most often, the H_2O molecule is used as the test molecule, for which good databases of spectroscopic parameters have been developed (line center position, absorption cross-sections, collisional broadening cross-sections). The choice of the H_2O molecule as the test molecule was dictated by the fact that, for diagnosing combustion processes in sub- and supersonic flows, the concentration of water vapor in the combustion zone is a key parameter characterizing fuel utilization efficiency, since the hydrocarbon components of the fuel are converted to water vapor during combustion. Additionally, there is a rich lineup of diode lasers developed for telecommunications applications in the IR range ($0.9-3\,\mu$m), where sufficiently strong absorption lines of the H_2O molecule are located.

The CO_2 molecule is also successfully used for diagnosing gas parameters, as it is produced in large quantities during combustion [12]. his molecule has strong absorption lines in the range above $2.7\,\mu$m. The authors' choice of the water molecule for diagnostics was dictated solely by the availability of a set of relatively inexpensive distributed-feedback (DFB) diode lasers and photodetectors at their disposal.

It should be noted that most developed DLAS variants are well suited for objects with uniform temperature distributions, as realized in laboratory furnaces, supersonic jet tunnels, or special model objects. The task of DLAS diagnostics becomes substantially more complex for media with non-uniform distributions of temperature and partial pressure of absorbing water molecules. In real power plants, the distributions of temperature and concentration are always non-uniform, at least due to near-wall boundary effects in combustion chambers. The method for reconstructing the temperature and concentration profiles in such systems remains a key and still unsolved problem.

The most general approach to solving the profile reconstruction problem is the tomographic method, which is well developed for X-ray computed tomography. In practice, this method has been demonstrated using special laboratory objects such as laminar flames from gas burners [13,14]. However, the tomographic method has not been applied in combustion chambers to date and is unlikely to be applicable due to the small volume of the chambers and the impossibility of introducing a large number of probing laser beams that intersect perpendicularly into such a volume.

For diagnosing such objects, typically only two small windows are used for input and output of the radiation — and the profile reconstruction problem is formulated in line-of-sight geometry, i.e., profile reconstruction from integrated absorption measurements along the entire path of the probing beam. As the DL probing beam passes through a non-uniform medium, the absorption line shape of the test molecule is the result along the entire probing path, including the path from the radiation source to the object and from the object to the detector. If measuring the integrated absorption of two lines with different lower-level transition energies is sufficient to determine the temperature of a uniform medium, then in a non-uniform medium in line-of-sight geometry, the number of parameters needed to characterize the object increases, requiring an increase in the number of recorded absorption lines. It should be noted that the most reliable DFB diode lasers can be rapidly tuned in frequency over a relatively narrow range. Therefore, the need to measure the intensities of a large set of absorption lines of the test molecule inevitably leads to an increase in the number of diode lasers used, and hence to an increase in the cost of the DLAS sensor and complication of the experimental spectrum processing procedure.

For developers of new aircraft engines, the most important parameter is the maximum temperature in the combustion zone, which determines fuel utilization efficiency. In this case, precise determination of the temperature distribution in various combustion zones is not critically important. For such a problem formulation, medium diagnostics can be performed using a smaller number of absorption lines.

The aim of this work is to determine the minimal set of absorption lines necessary for estimating the maximum temperature of a spatially non-uniform gas medium in single-beam line-of-sight geometry.

Selection of Absorption Lines

Problem Statement

In real diagnostics of a hot object, the intensities and shapes of selected absorption lines of the test molecule are measured experimentally, the experimental shapes are fitted with theoretically simulated ones based on spectroscopic databases, and the temperature is determined in the process of fitting the experimental profiles of all lines.

As noted above, solving the problem of reconstructing the temperature profile of a non-uniform medium requires measurements of absorption on a significantly larger number of test molecule lines [15,16]. This automatically increases the number of variables and substantially complicates not only the design of the DLAS sensor but also the mathematical algorithm for fitting experimental spectra with theoretical ones, since solving the inverse problem of spatial profile reconstruction becomes incorrect.

A detailed study of the number of lines required to reconstruct an arbitrary temperature distribution in the probed object was conducted in [17,18]. The study was based on a theoretical analysis of reconstructing the temperature distribution in the probed object from integrated absorption lines with known lower-level transition energies E''. It was found that the most accurate reconstruction of the

Table 1. Lower transition levels and line center frequencies

Number	1	2	3	4	5	6	7	8
E'', cm^{-1}	23.8	79.5	136.8	173.4	224.8	447.3	586.2	744.2
ν_c, cm^{-1}	7294.1	7306.7	7327.7	7343.8	7339.8	7368.4	7381.6	7393.8
Number	9	10	11	12	13	14	15	16
E'', cm^{-1}	782.4	1045.1	1327.1	1557.8	1806.7	2073.5	2660.9	2981.4
ν_c, cm^{-1}	7420.	7185.6	7426.14	7435.6	7444.37	7452.41	7466.33	6836.67

temperature distribution can be achieved by recording 14–15 lines uniformly distributed in energy E''.

For developers of new engines, determining the maximum temperature in a non-uniform medium is of greater interest than reconstructing the actual spatial profile of the combustion zone. In this case, a simpler trapezoidal temperature distribution can be assumed, with a quasi-constant maximum temperature in the central part of the object and a linear temperature drop toward its edges. In our experiments at the Zhukovsky Central Aero-Hydrodynamic Institute (TsAGI), such a profile was realized on a test bench. A similar distribution was considered in [19]. This simplified yet practically important model of a non-uniform medium allows reducing the number of lines needed to estimate the maximum temperature in a gas object to four lines, corresponding to the number of unknowns varied during fitting: maximum and minimum temperatures, molecule concentration, and the spatial fraction of the declining part in the trapezoid.

To determine the maximum temperature with its trapezoidal distribution, we developed a special modification of the direct absorption method without modulation, which enabled recording absorption below 10^{-2} against strong noise. The aim of this work is to describe the algorithm for selecting the optimal set of absorption lines for determining the maximum temperature of a gas object and to evaluate the error in determining this temperature for various line set variants. The development of new devices and experimental validation of the new DLAS sensor variant are the subject of a separate publication.

Calculation of Optimal Line Sets

A program was created to select optimal water molecule absorption lines that ensure minimal error in determining the maximum temperature in a non-uniform medium. Line selection was performed in the following sequence. First, in the wavelength range 1.3–1.6 μm the strongest lines with lower-level energies ranging from 0 (ground state of the H_2O molecule) to 3000 cm^{-1} were selected. Line selection was based on two criteria: minimization of interference from neighboring weak lines and maximum integrated intensity. Selection was performed using the HITRAN-2020 database. The 16 lines selected in this way are listed in Table 1.

Then, from these 16 lines, arbitrary combinations of 4 lines were formed, totaling 933 variants. Using any four arbitrary lines, the maximum temperature can be reconstructed with varying accuracy. To identify the optimal line set providing the best accuracy, we conducted a numerical experiment in which medium parameters were specified, and these parameters were determined by solving the inverse problem. The optimality criterion was the minimal error in reconstructing the a priori specified parameters. The calculations used the following object model: optical path length 12 cm, of which 10 cm is a section with constant maximum temperature, 2 cm is a linear drop to the minimum window temperature in the test chamber (500 K), water vapor concentration 0.05 atm (5%). Calculations were performed for two maximum temperature values 2200 and 1500 K.

The integrated absorption of lines for the selected object model was determined using the equation

$$A_i = S_i(T_1)P_{\mathrm{H_2O}}L_1 + \int_0^{L_2} S_i(T(x))P_{\mathrm{H_2O}}dx, \qquad (1)$$

where A_i [cm^{-1}] is the integrated absorption of the i-th line, S_i [cm^{-2}atm^{-1}] is the integrated intensity (line strength) of the i-th line normalized to the test molecule at 1 atm, T_1 is the maximum temperature of the model, L_1 is the length of the section with maximum temperature, L_2 is the length of the linear temperature drop section $T(x)$ from maximum to minimum temperature, and $P_{\mathrm{H_2}}$O is the test gas pressure (atm). Note that A is the integrated absorption at low absorption values. Otherwise, A is the integrated optical thickness (absorbance).

Using the HITRAN 2020 database and equation (1), the integrated absorptions of the lines A_i are found for the specified model parameters. Random values $\sim 10^{-5}$ cm^{-1} are added to the found A_i values to simulate experimental noise (white noise with SNR on the order of hundreds), and the inverse problem of reconstructing the model parameters is solved using the following algorithm.

By varying the parameters ($T_{\max}$, $T_{\min}$, L_2concentration), the current integrated absorptions of the four lines A_i are calculated. The best match of the found model parameters with the a priori specified ones is found using the nonlinear least-squares method. The added „noise“ leads to errors

Table 2. Error (σ(K)) in determining maximum temperature of 2200 K for five line combinations, line numbers from Table 1

T_{cal}	σ (K)	Line combination
2198.4	20.1	1, 6, 13, 16
2198.1	20.2	1, 7, 13, 16
2198.3	20.5	2, 6, 13, 16
2198.0	20.6	2, 7, 13, 16
2198.0	21.1	3, 7, 13, 16

Table 3. Error (σ (K)) in determining maximum temperature of 1500 K for five line combinations, line numbers from Table 1

T_{cal}	σ (K)	Line combination
1499.4	6.3	1, 6, 12, 16
1499.4	6.4	1, 7, 13, 16
1499.3	6.4	2, 6, 12, 16
1499.4	6.5	2, 6, 13, 16
1499.3	6.5	1, 7, 12, 16

Table 4. Error (σ (K)) in determining maximum temperature of 2200 K for five line combinations, line numbers from Table 1

T_{cal}	σ (K)	Line combination
2200.6	9.0	2, 10, 16
2200.6	9.0	3, 10, 16
2200.5	9.1	1, 10, 16
2200.6	9.4	5, 10, 16
2200.5	9.4	4, 10, 16

Table 5. Error (σ (K)) in determining maximum temperature of 1500 K for five line combinations, line numbers from Table 1

T_{cal}	σ (K)	Line combination
1500.3	2.4	1, 10, 6
1500.3	2.4	2, 10, 16
1500.3	2.4	3, 10, 16
1500.3	2.5	4, 10, 16
500.2	2.5	2, 8, 16

in determining the model parameters. The results of these calculations for two maximum temperature values are given in Tables 2 and 3. Based on these calculations, five combinations from the 933 line sets were selected that ensure minimal error. In the tables below, the temperature obtained from the described procedure is denoted T_{cal}.

It should be noted that the optimal line sets for maximum temperatures of 1500 and 2200 K are similar. The root-mean-square error in determining the maximum temperature varies insignificantly for different line sets. In diagnostics of flames in stationary burners, it is often assumed that, unlike in combustion chambers, the water vapor concentration is proportional to temperature. In this case, fitting experiments with a model of constant water vapor concentration underestimates the temperature by approximately 40 K.

The availability of commercial wall temperature sensors can allow determination of the actual low temperature (e.g., 500 K). In this case, three lines (three lasers) suffice to determine the maximum temperature. Optimal line sets for this case are given in Tables 4 and 5.

Analysis of the data in Tables 4–5 shows that in all cases, to minimize the error in determining the high temperature, lines with both low and high lower-level energies must be used. The maximum absorption for some of these lines in the model object does not exceed 10^{-2} at a typical water vapor concentration of 0.05 atm (5%) and temperature of 2200 K. This means that for practical implementation of a possible profile reconstruction algorithm, it is necessary to enable absorption measurements under conditions of strong interference at the level of order 10^{-3}.

The procedure for determining integrated line absorptions was described above. However, in a real experiment, integrated absorptions are determined from recorded spectra. In the numerical experiment described, for the object model used, absorption spectra in the vicinity of the selected strong lines were calculated using the formula

$$\alpha(\nu) = \sum_j S_j(T_1) g_j(\nu - \nu_j) P_{\mathrm{H_2O}} L_1 + \sum_j \int_0^{L_2} S_j(T(x)) g_j(\nu - \nu_j) P_{\mathrm{H_2O}} dx, \qquad (2)$$

where index j corresponds to a line from HITRAN2020 in the specified spectral interval, $g_j(\nu)$ is the shape of the j-th line. In calculating absorption spectra using formula (2), integration is replaced by summation with a step of 1 K.

Examples of spectral sections calculated in this way with lines 1, 6, 13, 16 are shown in Fig. 1. These spectra were calculated for the medium parameters noted above and specified maximum temperatures of 2200 or 1500 K at a water vapor concentration of 0.05 atm (5%).

In the numerical experiment described, the simulated spectra are treated as „experimental". At the first stage, the integrated absorption of the selected lines must be determined from these spectra.

This is usually achieved by fitting simulated spectra to experimental spectra using nonlinear least squares, with line profiles fitted by Voigt shapes. The Voigt shape well matches line shapes for uniform temperature and concentration distributions. However, in our case, the temperature is non-uniform. In this situation, the authors use a hybrid line fitting method: first, the line wings are fitted with Voigt (the central part of the line is not included in the fit) [15,16]. Then, numerical integration determines the fraction of integrated absorption in the line center, given by the difference between the experimental

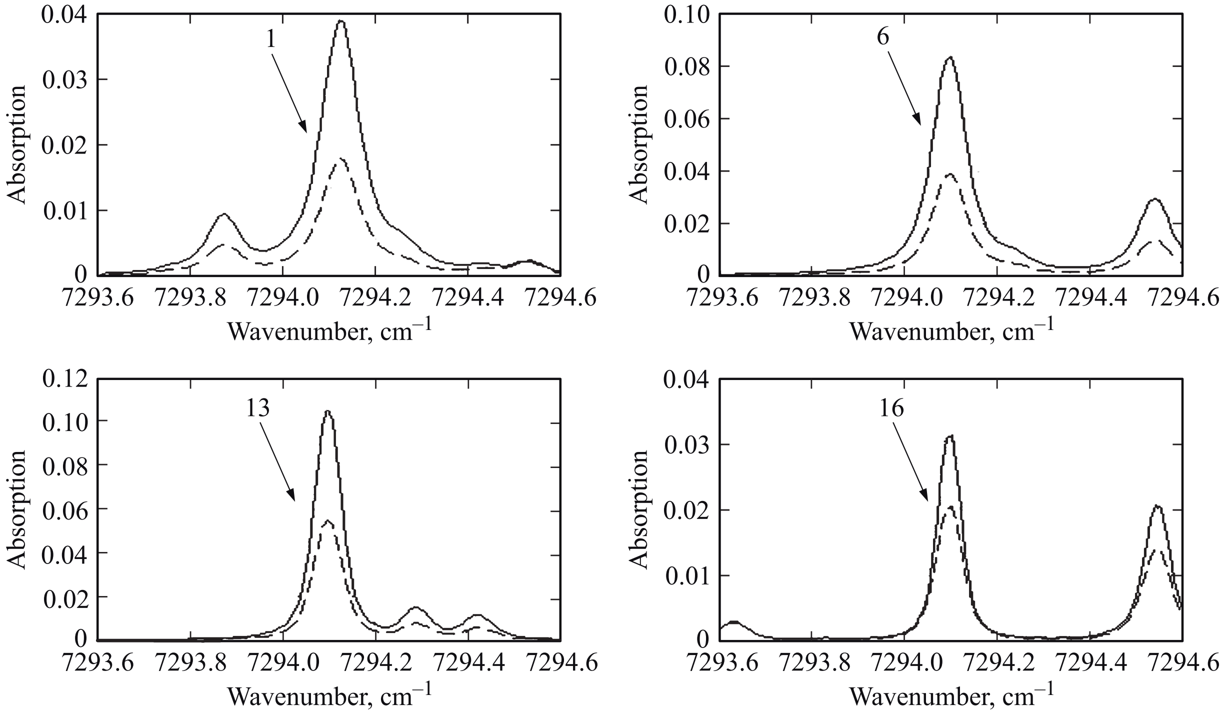

Figure 1. Absorption spectra on lines 1, 6, 13, 16. Lines are marked with arrows. Solid line — high temperature 1500 K, dashed line — 2200 K.

Table 6. Results of calculations using two line shape approximation methods

Variant	$T_{\min}$	$T_{\max}$	Declining section temperature length, cm	Water vapor pressure, atm
2 Voigt	498.77	1503.74	2	0.0502
2 Voigt	499.01	2201.78	1.98	0.0503
Hybrid	489.06	1494.3	1.87	0.05
Hybrid	492.34	2193.96	1.92	0.05

line profile and the simulated Voigt profile. The total integrated line absorption is then the sum of this difference and the integrated absorption of the simulated Voigt profile. This hybrid method requires numerical calculation of the difference, which can introduce errors in determining the integrated line absorption under high noise conditions in a real experiment.

We propose using fitting with two Voigt contours to determine integrated absorption. The line's integrated absorption is the sum of the integrated absorptions of these two contours. Knowing the integrated absorptions of the lines, for each combination of four lines, the inverse problem of calculating the model parameters is solved. We present actual results of such calculations for these two approaches using the example of the object model considered, taking lines 1, 6, 13, 16 without accounting for noise and interference from neighboring weak lines. The calculation results are listed in Table 6.

The results are similar. The algorithm we propose for calculating integrated intensity using two Voigt contours is technically simpler, as it does not require numerical calculation of the difference used in the hybrid method [15,16]. Note that solving the inverse problem of reconstructing model parameters leads to errors in both methods even assuming no noise and no interference from neighboring weak lines. This is due to inaccuracies in reconstructing line contours (real experimental or „model“) using both the hybrid method and the two-Voigt method.

In real spectra near strong lines, there are always nearby weaker lines whose wings nevertheless contribute to the integrated contour of the strong line. When fitting actually recorded spectra, the influence of such weak neighboring lines must be accounted for. This increases the number of fit parameters (due to the integrated intensities of weak lines) and leads to instability in the fitting procedure. To improve algorithm stability, we propose parametrically linking the intensities of weak lines to the intensities of neighboring strong lines in a specific spectral interval. For this, during Voigt profile fitting, the temperature and concentration of the test molecule are used as auxiliary

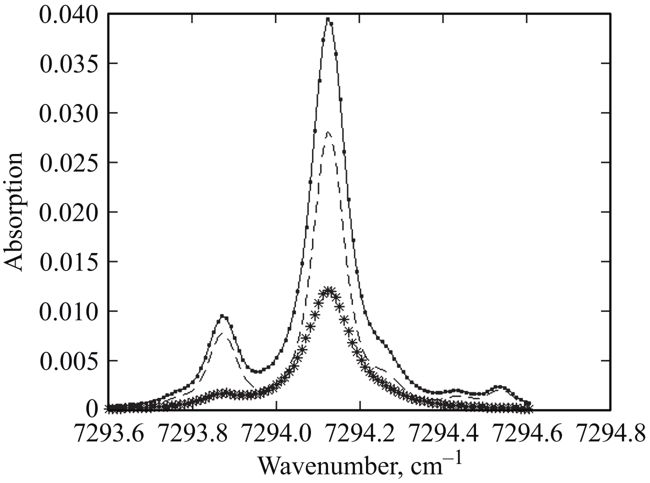

Figure 2. Absorption spectrum near 7294 cm^{-1}. Solid line — absorption spectrum in the 7294 cm^{-1} range, black dots — fit spectrum (overlaps with solid line), stars — contribution of low-temperature Voigt contour, dashed line — contribution of high-temperature Voigt contour.

Table 7. Results of model parameter calculations for lines 1, 6, 13, 16 with two-Voigt line fitting for two maximum temperatures

$T_{\min}$	$T_{\max}$	Declining section temperature, cm	Water vapor pressure,atm
491.94	1495.5	1.93	0.0499
487.86	2180.31	1.91	0.0495

fit parameters. Individual fit parameters are selected for each spectral interval. Thus, the intensities of weak lines become consistent with the intensities of strong lines in the considered spectral range rather than independent. An example of fitting a spectral section in the 7294 cm^{-1} range (line 1 in Table 1) with two Voigt contours is shown in Fig. 2.

Note that the spectral section synthesized using formula (2) is well fitted by the proposed two-Voigt method.

Verification of the developed algorithm was performed in several stages. Initially, model parameters ($T^0_{\max} = 1500$ K or 2200 K, $T^0_{\min} = 500$ K, $L^0_2 = 2$ cm, $P_{\mathrm{H_2O}} = 0.05$ atm) were selected, and using HITRAN 2020 data, four spectral sections near lines 1, 6, 13, 16 were simulated, which are treated as „experimental“ in subsequent calculations. Using the proposed two-Voigt fitting method, the „experimental“ integrated intensities of the selected strong lines were determined. At the next stage, the inverse problem of finding the model parameters was solved. For this, the set of parameters providing the best fit of the integrated intensities of the four lines, determined using formula (2), to the „experimental“ values was found using nonlinear least squares. The results of this verification are given in Table 7.

To simplify the DLAS sensor design by reducing the number of lasers required for accurate maximum temperature estimation, a search was conducted for water molecule line pairs that can fall within the rapid current tuning range of a single DFB laser (~ 3 cm^{-1}). Line selection is based on several criteria. First: the intensity of lines with similar lower levels must be at least 10% of the maximum line intensity. Second: the distance between lines must exceed 0.3 cm^{-1}, but be less than 1 cm^{-1}. Third: the difference in lower-level energies must exceed 400 cm^{-1}. Fourth: the intensity of weak lines does not exceed 0.02 of the intensity of the maximum line in the range. Using these criteria, only one pair of lines was found that allows using two DLs to record absorption on four lines:

1) DL1 —lines 7164.901 cm^{-1} (E 1394.81 cm^{-1}) and 7165.821 cm^{-1} (E 212.16 cm^{-1}),

2) DL2 lines 7358.741 cm^{-1} (E 2919.63 cm^{-1}) and 7359.33 cm^{-1} (E 1394.814 cm^{-1}).

Fig. 3 shows the spectral intervals containing the lines (marked with arrows) used to determine the high temperature.

We note immediately that restricting line selection to a narrow spectral range leads to degraded results. When determining the high temperature using these line pairs, the error increases by almost a factor of 4 compared to the case above with four lasers operating in different spectral ranges. The increased error is due to the fact that line pairs within the tuning range of one DL (~ 3 cm^{-1}) are generally weaker.

Conclusion

The features of diagnosing parameters of spatially non-uniform gas media in the single laser beam probing variant (line-of-sight) were investigated. In this geometry, the spectral contour of an absorption line is the resultant along the entire laser beam path, and the task of determining integrated absorption is mathematically ill-posed. For the best reconstruction of the spatial temperature profile, absorption measurements on 16 spectral lines are necessary. The task simplifies for the important case of determining the maximum temperature assuming a trapezoidal dependence of medium parameters from the maximum to the boundary. In this case, 4 absorption lines suffice. A program was created to select the optimal set of absorption lines of the test molecule H_2O in the spectral range $1.3-1.4\,\mu$m. Various combinations of H_2O absorption lines were found that enable determination of the maximum temperature. A modified variant of fitting integrated absorption lines in a spatially non-uniform medium using two Voigt contours was proposed. An assessment of the accuracy of maximum temperature determination for various line sets was performed for a specific experimental situation (optical path length 12 cm, of which 10 cm is a section with constant maximum temperature and 2 cm is a linear drop to the minimum

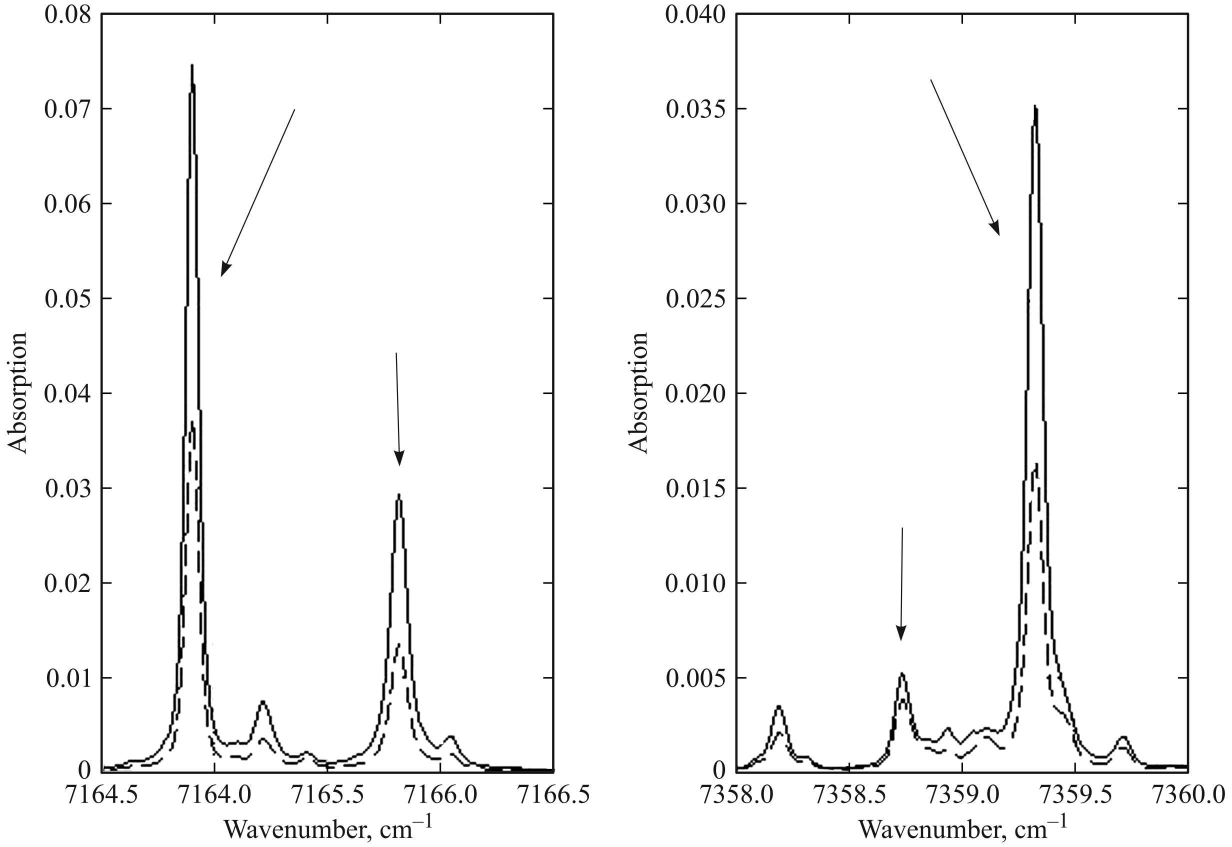

Figure 3. Solid graph — high temperature 1500 K, dashed graph — high temperature 2200 K, strong lines marked with arrows.

window temperature in the test chamber). The error assessment for determining temperature was conducted for maximum temperatures of 2200 and 1500 K at a minimum temperature of 500 K. The errors in determining these maximum temperatures Δ are approximately 20 K for 2200 K and 10 K for 1500 K.

An assessment was conducted for the experimental variant where the cold boundary temperature of the tested gas object (windows) is not an unknown varied parameter but is measured by a thermocouple. In this case, only three diode lasers operating in different spectral ranges can be used. The accuracy of maximum temperature determination improves by approximately a factor of2−3 in this case.

For the case using only two DLs, within whose tuning ranges multiple lines fall, the developed algorithm for reconstructing the maximum temperature of a spatially non-uniform gas medium can be used. However, for this case, the temperature determination error increases substantially, which is explained by the non-optimal set of absorption lines located in the limited DL tuning interval.

Conflict of interest

The authors declare that they have no conflict of interest.

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