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RECONSTRUCTION OF N_2O AND CH_4 CONTENT BY DIAL MEASUREMENTS AT WAVELENGTHS OF OVERTONE CO LASER

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ABSTRACT

The paper presents the results of laboratory experiments on measurement of absorption and extinction of radiation of the overtone CO laser at wavelengths used for sensing of methane and N_2O in the mid-IR spectral range with the differential absorption (DIAL) method, as well as the concentrations of the studied gases reconstructed from the analysis of experimentally obtained absorption coefficients.

1. INTRODUCTION

In connection with the growing pollution of the atmosphere, the problem of real-time monitoring of gas concentrations becomes increasingly urgent. This problem can be solved with the aid of remote laser sensing of the atmosphere. The maximal information in remote determination of the atmospheric composition with high temporal resolution can be obtained only by the optical method with the use of lasers, that is, by lidar method [1, 2]. The development of laser remote IR spectroscopy requires the development and implementation of new laser sources in the mid-IR spectral region generating radiation in the as wide as possible spectral range with a small frequency step. The tuning range of the overtone CO laser (2.5-4.2 µm) [3, 4] allows investigations in the field of laser spectroscopy, as was demonstrated in [5 - 11]. In addition to the laboratory laser spectroscopy, the overtone CO laser is interesting for some applications, which require the transport to long distances, because the range of the overtone radiation covers the atmospheric transparency window in the spectral range 3.3-4.2 µm [1]. The absorption of radiation of the overtone CO laser in the atmosphere was studied in [12, 13], where both the linear and nonlinear absorption, including induced absorption and "bleaching" by laser radiation, were taken into account in the calculations. The list of 50 spectral lines of the overtone CO laser characterized by the lowest absorption in the atmosphere is given in [12]. The minimal absorption coefficient at such lines in the atmosphere is about 0.01 km^{-1} (H_2O and CO_2 continuum absorption taken into account). At the same time, the overtone CO laser can find application in remote sensing of the atmosphere. The weak absorption of laser radiation in the atmosphere allows spectroscopic measurements of concentrations of various substances at long distances [14]. It was demonstrated in [15, 16] that lines of the overtone CO laser are promising for remote analysis of minor gas constituents (MGCs) of the atmosphere.

The aim of this study was to conduct model laboratory experiments on remote measurement of absorption and total extinction of radiation of the overtone CO laser in methane and N_2O with the following reconstruction of concentrations of the minor gas constituents under study.

2. EXPERIMENT

Laboratory experiments on lidar sensing of methane and N_2O by the path DIAL scheme were carried out at the wavelengths selected by the technique developed in [16] for sensing of methane (3.440 µm) and N_2O (3.877 µm). The experiments were conducted at the laser setup developed in the Laboratory of Gas Lasers of the Lebedev Physical Institute RAS [17].

The absorption and extinction of radiation scattered from a topographic target and passed through the medium with the studied gas were measured with OPHIR 3A-SH calorimeters and FSG-22-3A1 microcryogenic Ge:Au photodetectors (SVOD photoresistors). The optical arrangement of experiments on measurement of the absorption and extinction coefficients of radiation of the overtone CO laser in methane and N_2O is shown in Fig. 1.

The laser cavity of the overtone CO laser was formed by the M1 spherical mirror (R~20 m) and the diffraction grating (420 lines/mm, blazing angle of 27°) operating in the autocollimation

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mode in the first diffraction order and outputting the radiation in the zero diffraction order. The laser beam aperture was determined by the D1 intracavity diaphragm 25 mm in diameter. The He-Ne laser was used for alignment of the laser cavity. The spectral tuning was performed by turning the diffraction grating. The grating turning angle was controlled with an auxiliary semiconductor laser (LTs-1, radiation wavelength of 0.65 μm).

In this configuration, a 10-cm long cell was filled with the gas mixture at a pressure of 1 atm: studied gas constituents with nitrogen (N_2O:N_2, CH_4:N_2) in proportion 1:24 at the 4% concentration of the absorbing gas. To measure the laser pulse energy E_0, a part of the laser radiation passed through the diaphragm D2 20 mm in diameter was directed to the first calorimeter through reflection from the ZnSe plane-parallel plate and the spherical mirror M2 (R~0.25 m).

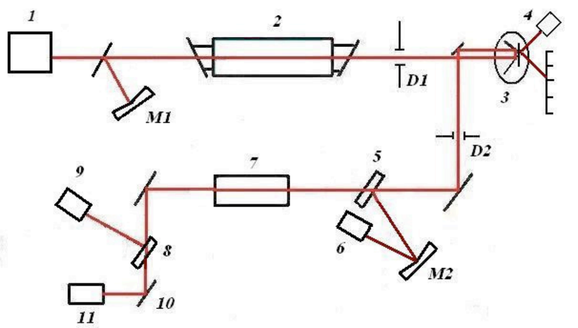

Figure 1. Experimental setup for measurement of absorption and extinction of radiation scattered from the topographic target and passed through the medium with the studied gas: (1) He-Ne laser, (2) CO laser, (3) diffraction grating, (4) semiconductor laser, (5) ZnSe plate, (6) Ophir 3A-SH calorimeter, (7) absorbing cell (L=10 cm), (8) CaF_2 plate, (9) IRS/ Ophir 3A-SH calorimeter, (10) topographic target, and (11) SVOD photoresistor

As the laser radiation passed through the cell, a part of the radiation was directed to the second calorimeter /IRS (infrared spectrometer) through reflection from the CaF_2 plane-parallel plate. The second calorimeter measured the energy of the laser beam E passed through the absorbing cell. The absorbed radiation was calculated in accordance with the Bouguer—Lambert—Beer law. Another part of the laser radiation was directed to the topographic target behind the CaF_2 plate. The topographic target was represented by the diffusely scattering reflector with an albedo of 0.8. The SVOD photoresistors measured the energy of radiation passed through the medium with the studied gas and the radiation part lost at reflection from the topographic target. The radiation extinction was measured by the technique described in [16]. The calculated and measured absorption and extinction coefficients for N_2O and methane are summarized in Table 1.

Table 1. Calculated and measured absorption and extinction for N_2O and methane

λ in air, μm	abs. coeff., cm^{-1} (calculation)	abs. coeff., cm^{-1} (experiment)	extinction coeff., cm^{-1} (experiment)
	N_2O		
3.841	0.0014	0.0095	-
3.852	0.0067	0.0004	0.0129
3.877	0.1178	0.0953	0.1202
3.892	0.0209	0.0502	-
3.897	0.0102	0.0072	-
3.907	0.0508	0.0262	0.0029
3.918	0.0307	0.0223	0.0066
3.930	0.0061	0.0014	0.0185
	CH_4		
3.328	0.0025	-	0.0090
3.336	0.0005	0.00462	0.0151
3.345	0.0381	0.0199	0.0276
3.432	0.0003	0.0029	-
3.440	0.0247	0.0078	0.0564
3.444	0.0011	-	0.0563
3.453	0.1159	0.1040	0.1733

It can be seen from the tabulated experimental data on absorption and extinction that the measured (absorption and extinction) and calculated (absorption) values are in a good agreement in the entire spectral range, in which the measurements were conducted. Some discrepancies between the calculated and measured methane absorption coefficients can be explained by the influence of residual interfering absorption of water vapor in the measuring cell or (for calculation of the extinction coefficient) by the inhomogeneity of the laser radiation scattered from the topographic target and passed through the medium with the gas mixture under study. The error of energy measurement by the OPHIR 3A-SH calorimeter, which did not exceed 5%, should be taken into account as well.

3. RECONSTRUCTION OF THE GAS CONCENTRATIONS

For the wavelengths selected for sensing of methane and N_2O at the operation with the topographic target, the inverse problem of reconstruction of the concentrations of this minor atmospheric gases in the 10-cm long absorbing cell with the aid of the overtone CO laser was solved from the analysis of the experimentally determined absorption coefficients. The reconstructed concentrations in comparison with the concentrations in the calibration mixture of the studied gases in the cell are shown in Fig. 2.

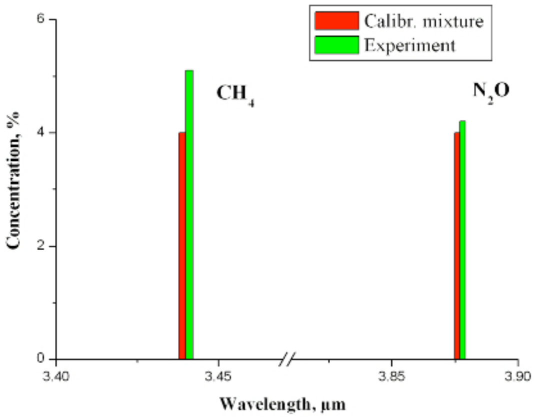

Figure 2. Reconstructed concentrations of N_2O and methane in the case of overtone CO laser radiation at the selected sensing wavelengths

It can be seen from the figure that the accuracy of measurement of the absorption coefficients of the gases under study influences significantly the solution of the inverse problem of concentration reconstruction.

4. CONCLUSIONS

The test laboratory experiments (based on numerical simulation) on measurement of absorption and extinction of the overtone CO laser radiation in mixtures with the studied gases yield the acceptable agreement between the calculated and measured values except for few noninformative wavelengths, which allows the developed technique of wavelength selection to be applied along with the DIAL method. The reliability of the obtained results is confirmed by the solution of the inverse problem on reconstruction of the concentrations of the studied gas constituents from analysis of the experimentally obtained absorption coefficients.

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