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REMOTE SENSING OF ATMOSPHERE, HYDROSPHERE, AND UNDERLYING SURFACE

Remote Analysis of Methane Concentration in the Atmosphere with an IR Lidar System in the 3300–3430 nm Spectral Range

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Abstract—A differential absorption lidar (DIAL) system based on optical parametric oscillators (OPO) with nonlinear KTA and KTP crystals is designed. The crystals allow laser radiation tuning in the IR wavelength region. A series of experiments on remote monitoring of methane along a horizontal surface sounding path in the 3300–3430 nm spectral range was carried out. Based on the experimental results, the CH_4 concentrations are retrieved along a surface 800-m path in the spectral range under study with a spatial resolution of 100 m.

Keywords: lidar, infrared region, optical parametric oscillator, differential absorption, methane

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INTRODUCTION

Contemporary laser radiation sources used in the solution of atmospheric problems are designed on the basis of broadband IR molecular lasers and parametric frequency converters based on nonlinear crystals, which generate overtones, harmonics, and total and difference frequencies of laser radiation covering the spectral range from 2 to 18 μm. This range is the most promising for monitoring almost all atmospheric gases. The lasing spectra of existing lasers with energy parameters acceptable for lidar measurements cover only some parts of this range; therefore, the design of differential absorption lidar (DIAL) systems which allow controlling the entire range is an topical task.

Advances in the study and production of nonlinear optical crystals have led to the creation of ever new gas analysis systems [1–3] thereby expanding the capabilities of remote monitoring of the environment. For example, a DIAL system based on two β-BaB_2O_4 (BBO) optical power amplifiers pumped by an Nd:YAG laser with a pulse length of ~3 ns, a pulse repetition rate of up to 100 Hz and the tuning range 400–2500 nm is described in [4]. As experimental results, the authors present the vertical distribution of water vapor along a 1500-m path measured at wavelengths $\lambda_{on} = 1187.869$ nm and $\lambda_{off} = 1187.716$ nm with a spatial resolution of 15 m. The use of a $LiNbO_3$ crystal pumped by an Nd:YAG laser allowed the authors [5, 6] to implement a gas analysis system with the laser wavelength tuning ranges 1410–1850 and 2900–4100 nm, pulse energy of 1–45 mJ, and lasing line width of 3–3.5 cm^{-1}. The system specifications include a possibility of detecting CH_4 using topographic targets at a distance of 2–5 km, with a path-integral device sensitivity of 1 ppm.

A DIAL system based on a parametric light oscillator (OPO) with KTP and pumped by a Nd:YAG laser with an output energy of 70–100 mJ per pulse made it possible to measure daily variations in CO_2 at $\lambda =$ 1570 nm for the first time [7]. A laser source based on crystals with a periodic domain structure, which operates in the spectral range 3300–3700 nm, is presented in [8]. The authors report that the radiation source they developed allows simultaneous detection of H_2O and CH_4. One recent publication [9] provides more detailed information about a similar system with operating wavelengths near 2000 nm.

$LiNbO_3$ crystals with a periodic domain structure allow one to study the concentrations of such atmospheric gases as CO_2, CH_4, and H_2O. This is one of the steps in the implementation of a multicomponent gas analysis system based on a similar radiation source [9]. In [10], the authors presented a laser which is based on a Nd:YLF diode-pumped laser and KTP OPO. The laser allowed successful 2-km path-integral measurements of the CO_2 concentration. Further research allowed implementing a system suitable for in-field automatic detection of CO_2 and CH_4 in three dimensions [11].

Work [12] describes a unique mobile lidar designed for remote sounding of vapors of key substances used for the manufacture of explosives (acetone and nitromethane). Laboratory measurements with the use of

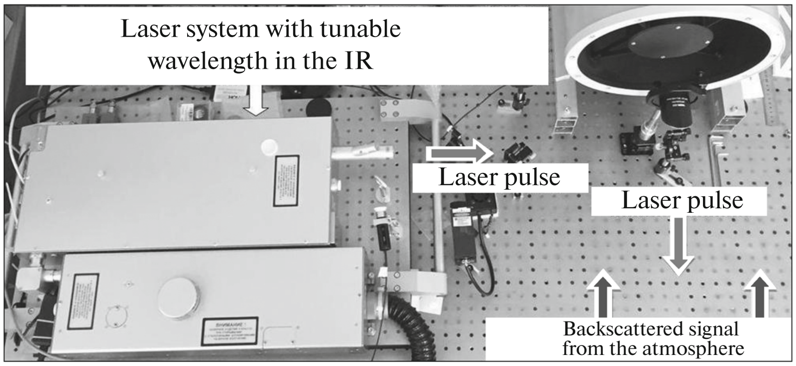

Fig. 1. IR DIAL system.

spectroscopic databases showed the spectral range 3000–3500 nm to be optimal for the detection of C_3H_6O and CH_3NO_2 vapors by the differential absorption method. The output laser pulse energy is 12 mJ, the beam diameter is 22 mm, the pulse length is 5.2 ns, the pulse repetition rate is 10 Hz, the wavelength tuning time is 0.3 s, and the lasing line width is about 5 cm^{-1}.

The active development of this field is confirmed by a large number of patents and publications devoted to the development of IR differential absorption lidars and techniques for atmospheric gas analysis, including a multichannel lidar with several OPOs [13], an aircraft-based lidar [14], a technique which combines differential absorption and frequency comb methods [15], a broadband tunable laser source [16–18], a lidar for sounding atmospheric methane in the range 3000–3450 nm [19], and lidars [20–22].

The aim of this work was the design of a system for remote monitoring of background and above-background methane concentrations along surface tropospheric sounding paths.

IR DIAL SYSTEM

An IR OPO DIAL system has been designed at the V.E. Zuev Institute of Atmospheric Optics, Siberian Branch, Russian Academy of Sciences (IAO SB RAS) for the study of distribution of atmospheric gases in the surface air layer (or in the lower troposphere) which significantly absorb in the mid-IR spectral region (3000–4000 nm). This range includes CH_4 absorption bands (3300–3430 nm), which allows one to study its total content in the lower troposphere.

Figure 1 and 2 show the IR DIAL system, its receiving telescope and mirror collimator. The specifications of the system are tabulated.

EXPERIMENTS AND MEASUREMENT RESULTS

The lidar system designed was used in a series of laboratory experiments on measurements of the laser radiation absorption by methane, which was a component of a molecular mixture. For the experiments, the spectral range 3300–3430 nm was chosen, since it includes a quite strong methane absorption band (on- and off-line sounding wavelength pairs were selected in that sounding range [23, 24]). We used a $CH_4 : N_2$ (2 : 98) molecular mixture in a cell at a pressure of 1 atm.

The optical scheme of the experiments is shown in Fig. 3.

An OPO laser, described in detail in [24–27], was used as a radiation source of the lidar system. The radiation from the laser system was directed to a KG-01 gas cell (100 mm long, 40 mm diameter CaF_2 windows) with the help of plane-parallel CaF_2 plates and a rotary mirror. Before propagating through the gas cell, a part of the radiation was guided onto thermally cooled photodiode *2* by means of reflection from a CaF_2 plate (split ratio was 50/50) to record the reference signal. Thermally cooled photodiode *3* measured an informative signal at the gas cell exit. The transmission spectra of the gas mixture were calculated from the ratio of the reference and informative signals. The coefficient of OPO laser radiation absorption by the molecular mixture in the gas cell was calculated by Bouguer's law.

The mirror collimator and receiving telescope make it possible to record backscattered radiation

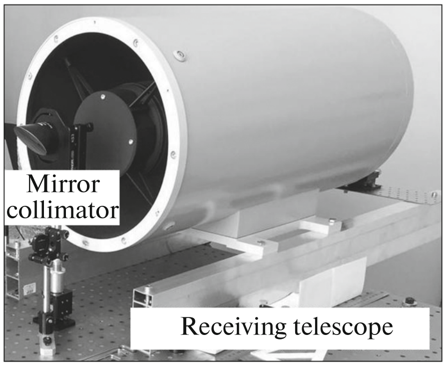

Fig. 2. Receiving telescope and mirror collimator.

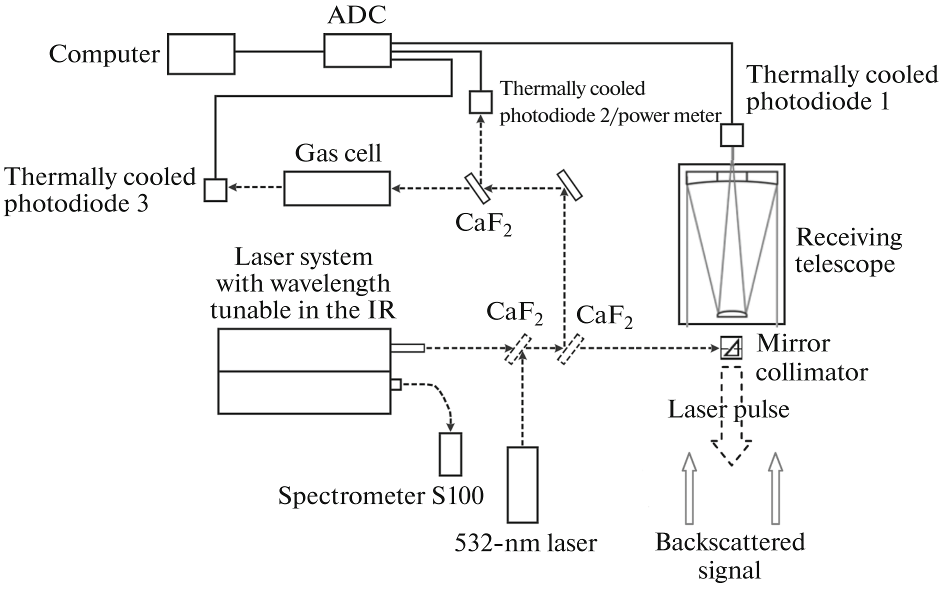

Fig. 3. Optical scheme of experiments with the IR DIAL system.

Table 1. Specifications of the IR lidar system

Parameter	Value
Laser	
Wavelength tuning range, nm	3300–3430
Maximal pulse energy, mJ	6
Lasing line width, cm^{-1}	1
Pulse length, ns	10–13
Pulse repetition range, Hz	10
Beam divergence, mrad	2
Mirror collimator	
Focal length of parabolic mirror 1, mm	15
Focal length of parabolic mirror 2, mm	152.4
Optical receiving system	
Type	Cassegrain
Effective focal length, mm	1457
Receiving aperture diameter, mm	300
Diameter of circle of confusion (at 532 nm), µm	70
ADC of signals	
Number of analog outputs	4
Transmission band, MHz	100
Resolution, bit	12
Sampling frequency over all channels, Gsample/s	1
Photo detector	
Spectral range, µm	1–4
Diameter of photosensitive area of photodiode, µm	300
Rise time, ns	50
Detectability D^*, cm $Hz^{1/2}$ W^{-1}	4×10^9

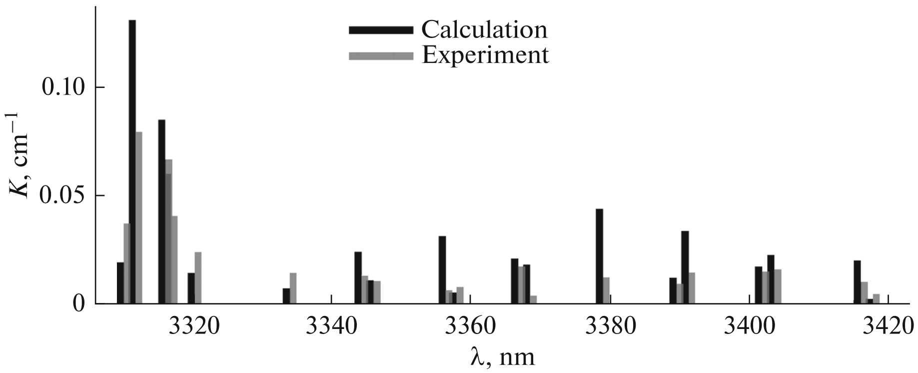

Fig. 4. OPO laser radiation absorption coefficient of the $CH_4 : N_2$ (2 : 98) molecular mixture at a pressure of 1 atm in the methane sounding informative wavelength range (3300–3430 nm) calculated and measured on December 13, 2018.

Fig. 5. Sounding radiation propagation direction.

from the atmosphere and to use the cell as a calibration component during field measurements. The mirror collimator consists of two parabolic mirrors with focal lengths of 15 and 152.4 mm, respectively; it allows reducing the laser radiation divergence. During atmospheric measurements, a part of the radiation is reflected from two CaF_2 plates to a power meter (PM) with the aim of controlling the output radiation power. The laser which operates at a wavelength of 532 nm is used to adjust the output radiation of the IR lidar system and to ensure the alignment of the axes of visible and IR radiation. A spectrometer is necessary to control the wavelength of the output radiation of the IR lidar system. Thermally cooled photodiode *1* records a backscattered signal from the atmosphere collected by the receiving telescope.

Figure 4 shows the OPO laser radiation absorption coefficients calculated with the use of the HITRAN database [28] and measured during the above-described experiments.

The tuning to more than 60 OPO laser wavelengths was performed in the spectral range under study (3300–3430 nm), including both informative and noninformative methane sounding wavelengths; the information about them can be useful for calibration of the OPO lidar system.

The gas mixture transmission spectra were measured by tuning the laser radiation wavelength. After processing the experimental data, the methane absorption coefficients were retrieved (Fig. 4), which confirmed the spectral position of the lidar emission lines, as well as the presence in the real atmosphere of the absorption lines of a gas under study presented in the spectroscopic databases.

The absorption coefficients can differ in the spectral regions noninformative for the gas analysis because of interfering absorption by external gases. The absorption coefficients measured (by the laser pulse energy after propagation through the gas cell) are in good agreement with the absorption coefficients calculated with the use of the HITRAN database at informative sounding wavelengths selected. It was decided to use the pair of sounding wavelengths 3415.711 (on-line) and 3417.484 nm (off-line) in further measurements of atmospheric CH_4.

The IR lidar system (see Fig. 3) was also used in field experiments on the study of CH_4 concentration distribution at the above sounding wavelength pair with a spatial resolution of 100 m (experiment geometry is shown in Fig. 5). The measurements were carried out in Tomsk on February 1, 2019.

Figure 6 shows the lidar signal received under wavelength tuning in the spectral range 3300–3430 nm (signals were alternatively accumulated at informative wavelengths at discrete sets of lidar system lasing lines; the

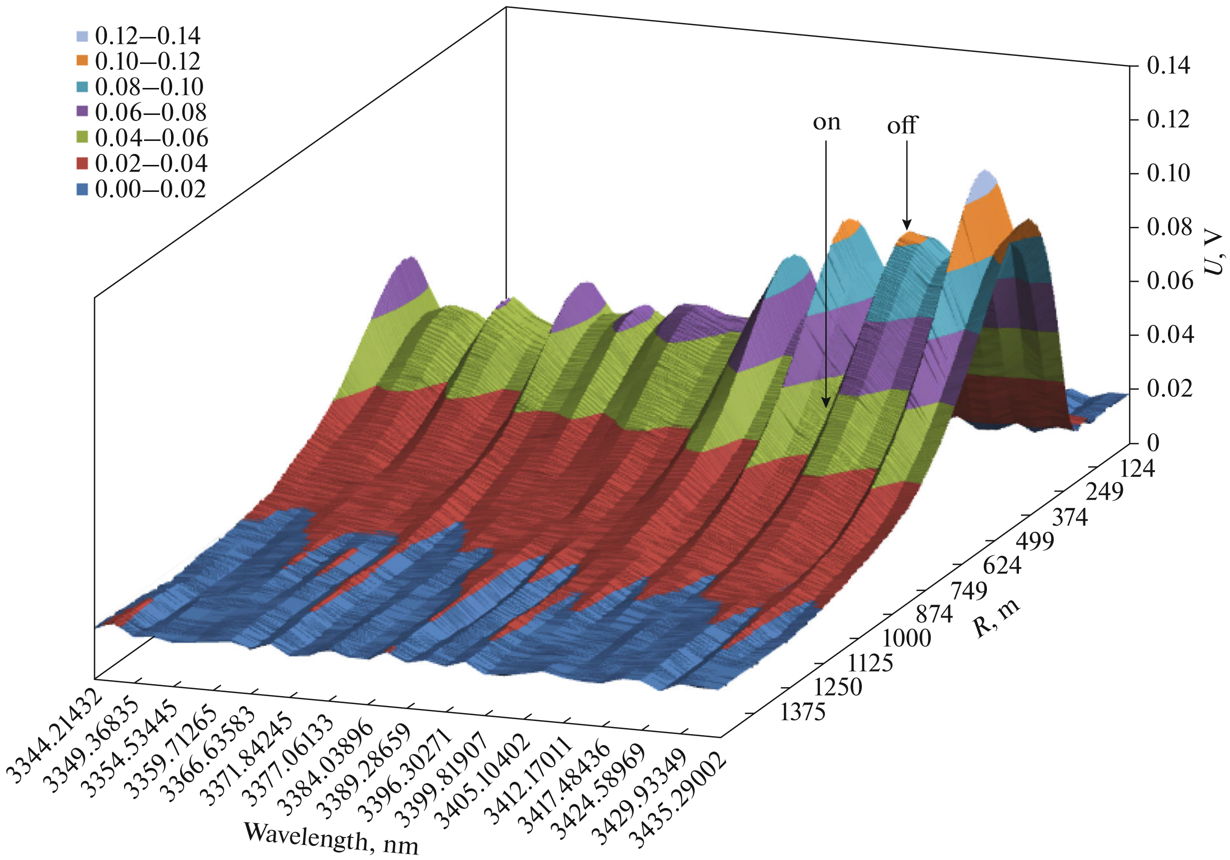

Fig. 6. Lidar signal received under wavelength tuning in the methane sounding informative range.

change time between the wavelengths was 35–50 ms, the averaging time was 6.4 s).

Figure 7 shows the CH_4 concentration distribution along a sounding path 800 m long, measured during the experiments, in comparison with the data obtained at the TOR station of the IAO SB RAS [29, 30], where the methane concentration is measured round-the-clock. The IR lidar system and the TOR station are located in the same building.

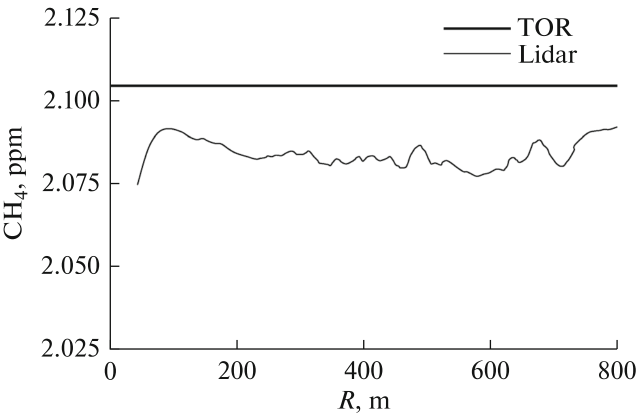

Fig. 7. CH_4 concentration measured along a 800-m surface air path with a spatial resolution of 100 m (February 1, 2019).

Figures 6 and 7 confirm a capability of the system designed for studying and monitoring the CH_4 concentration distribution in the surface air layer. The measurement results agree well with the data of numerical simulation and the TOR station data. They prove the system can be used for retrieving near-background and above-background CH_4 concentrations.

CONCLUSIONS

A DIAL system has been created that allows the reception and processing of backscattered IR signals at surface tropospheric sounding paths. The absorption of OPO laser radiation by methane was measured in laboratory conditions and in the real atmosphere. The experimental results show the capabilities of the lidar system designed to study and monitor methane concentration distributions along horizontal surface sounding paths up to 800 m long in the mid-IR range. The methane concentrations (~2.1 ppm) retrieved from the lidar signals are in good agreement with the TOR station measurements.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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