Доклади на Българската академия на науките
Comptes rendus de l'Académie bulgare des Sciences

Tome 76, No 3, 2023

PHYSICS
Atmospheric physics

DIAL SOUNDER OF METHANE INSUSCEPTIBLE TO VARIABLE HUMIDITY BASED ON BROADBAND LASER DIODES

Stoyan Penchev#, Vasilka Pencheva, Tanja Dreischuh

Received on November 16, 2022
Presented by P. Atanasov, Member of BAS, on January 31, 2023

Abstract

A novel differential absorption lidar method utilizing the spectral and energetic properties of pulsed, high-power, broadband laser diodes for remote sensing of methane is reported. The atmospheric gas content is retrieved using the derived calibration function, integrating multiple resonance lines of the selected $CH_4/Qy\nu_3$ second overtone absorption spectrum matched to a laser mode centred at optimal wavelength. The sensitivity of the lidar is effectively improved by multiplexation of the detected laser radiation in dual spectral channels eliminating the interference of the atmospheric water vapour.

Key words: greenhouse gases, methane, differential absorption lidar, laser diodes

Introduction. The high-symmetry molecule of methane is rich in vibration-excitation states transforming sunlight into heat for the global warming. Trends of growing emissions from industrial zones and melting [1,2]. Its content has doubled for 200 years that is responsible for the present acceleration of the greenhouse effect.

A lidar (light detection and ranging) is advantageous for retrieving range-resolved data of the atmospheric scattering. Particularly, a differential absorption

---

Corresponding author.

This research has been supported by the Bulgarian Ministry of Education and Science (support for ACTRIS BG, part of the Bulgarian National Roadmap for Research Infrastructure).

DOI:10.7546/CRABS.2023.03.03

lidar (DIAL) is capable to perform highly-selective simultaneous measurement of the atmospheric gas content by scattering and absorption. In the given research area, we contributed to pioneer studies advancing an effective DIAL technique by replacement of narrowband lasers with their broadband analogues [3–5]. Such broadband lidar detects the integral absorption spectrum of the gas instead of the amplitude of an isolated resonance line [6] that depends on pressure broadening. The absorption spectrum of methane around 1.66 µm wavelength is found feasible for DIAL sounding of its atmospheric content [5,7]. Also, the spectral band is favourable for the near-infrared (NIR) laser diodes and sensitive avalanche photodetectors which do not need cooling to low temperatures.

We have developed a DIAL method based on pulsed, high-power laser diodes of selected wavelengths that match confined segments of the monitored integral absorption spectrum (Fig. 1). However, methane spectrum centred at 1.667 µm is mixed with resonance lines of water vapour, unlike the particular spectral band around 0.91 µm used for DIAL hygrometer [3] that lacks interfering spectral features. An effective solution by multiplexation of the broad laser line eliminating the interference of water vapour in the detected signal is proposed. The resonance spectrum of $CH_4/Qy\nu_3$ rotational-stretching vibration is selected by investigation of its complex with CO_2 and H_2O absorption in 1.6–1.7 µm band using HITRAN (high-resolution transmission and molecular absorption), database of experimentally-derived and calculated parameters "for radiance calculations to predict and simulate the transmission and emission of light in the atmosphere" [8]. We also conducted a technological survey to verify that custom laser diodes as an extension of OSI LDI Inc. MCW563S-XXR and CVLL 350-CL90 commercial series towards 1.66 µm wavelength could be ordered [9].

Theoretical discussion. The integral resonance absorption function modulating the broad laser line over the lidar path is described by a convolution integral. The latter contains spectral terms of absorption lines of Lorentzian profile of methane and water vapour in the range of the laser line of Gaussian profile:

$$\begin{aligned}C &= \int_{\nu} f_{\mathrm{G}} \exp\left(-K\sum_{n} f_{\mathrm{L}}\right)\mathrm{d}\nu \\ &= \int_{\nu} \exp\left[-4\ln 2\left(\frac{\nu-\nu_{\mathrm{l}}}{\Delta\nu_{\mathrm{l}}}\right)^{2} - K\sum_{n} S_n \frac{\Delta\nu_{\mathrm{a}}^{2}/4}{(\nu-\nu_n)^{2}+\Delta\nu_{\mathrm{a}}^{2}/4}\right]\mathrm{d}\nu,\end{aligned} \tag{1}$$

where, $\Delta\nu_{\mathrm{l}}$ is laser linewidth; ν_n are central frequencies of the absorption lines; $\Delta\nu_{\mathrm{a}}$ is linewidth of the absorption lines at atmospheric pressure; S_n are absorption linestrengths and K is product of gas mixing ratio (GMR) and lidar path.

The exponent (Exp. 1) is approximated by difference for weak absorption, where step-functions β and $(1-\beta)$ taking values of unity and zeros that correspond

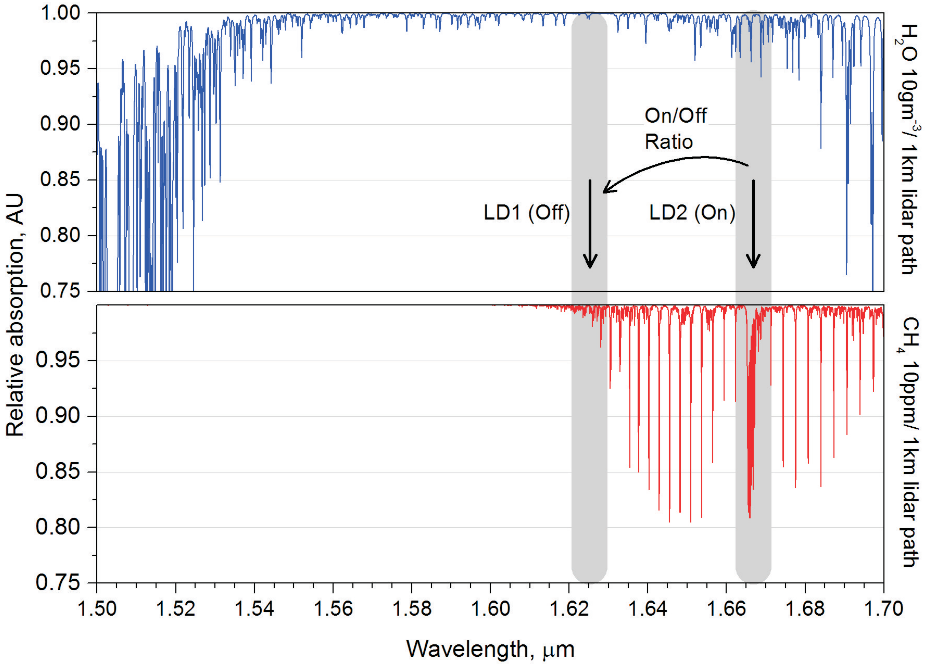

Fig. 1. Differential absorption λon/λoff bands matching selected CH_4 and H_2O NIR overtone spectra of relevant GMR and lidar path; shaded bars correspond to laser modes of 8 nm linewidth centred at 1.625 µm and 1.667 µm wavelength

to multiplexation in dual channels, using bandpass and notch filters:

$$(2)\qquad I_{\mathrm{DIAL}} = C_{\lambda\mathrm{on}}/C_{\lambda\mathrm{off}} \simeq \frac{\int_\nu \beta f_{\mathrm{G}}\left(1-K\sum_n f_{\mathrm{L}}\right)\mathrm{d}\nu}{\int_\nu (1-\beta)\, f_{\mathrm{G}}\left(1-K\sum_n f_{\mathrm{L}}\right)\mathrm{d}\nu},$$

$$(3)\qquad I_{\mathrm{DIAL}} \simeq \frac{\int_\nu \beta f_{\mathrm{G}}\left[1-\left(K\sum_n f_{\mathrm{L}}\right)_{\mathrm{H_2O}}-\left(K\sum_n f_{\mathrm{L}}\right)_{\mathrm{CH_4}}\right]\mathrm{d}\nu}{\int_\nu (1-\beta)\, f_{\mathrm{G}}\left[1-\left(K\sum_n f_{\mathrm{L}}\right)_{\mathrm{H_2O}}\right]\mathrm{d}\nu}.$$

The above expression is simplified further assuming equal small intensities of modulation of the laser line by water vapour in both spectral channels:

$$(4)\quad \int_\nu (1-\beta)\, f_{\mathrm{G}}\left[1-\left(K\sum_n f_{\mathrm{L}}\right)_{\mathrm{H_2O}}\right]\mathrm{d}\nu = \int_\nu \beta f_{\mathrm{G}}\left[1-\left(K\sum_n f_{\mathrm{L}}\right)_{\mathrm{H_2O}}\right]\mathrm{d}\nu$$

$$\int_{\nu} \beta f_{\mathrm{G}} \left(K \sum_{n} f_{\mathrm{L}} \right)_{\mathrm{H_2O}} \mathrm{d}\nu \ll \int_{\nu} \beta f_{\mathrm{G}} \mathrm{d}\nu. \tag{5}$$

The transformations result in a final expression of the DIAL signal, which is independent of absorption by water vapour.

$$I_{\mathrm{DIAL}} \simeq 1 - \frac{\int_{\nu} \beta f_{\mathrm{G}} \left(K \sum_{n} f_{\mathrm{L}} \right)_{\mathrm{CH_4}} \mathrm{d}\nu}{\int_{\nu} \beta f_{\mathrm{G}} \mathrm{d}\nu}. \tag{6}$$

The theoretical result is validated by spectral high-resolution data for determination of the optimal laser wavelength (next section).

Broadband DIAL sounder of methane. We advanced the DIAL method by optimization of the optical scheme to split the detected signal of differential absorption in dual spectral channels by means of a combination of beamsplitter and narrow bandpass and notch filters [10]. The optimal wavelengths and linewidths of the spectral filters are determined on the basis of HITRAN spectral data of molecular absorption for the modulated laser line assuming a Gaussian

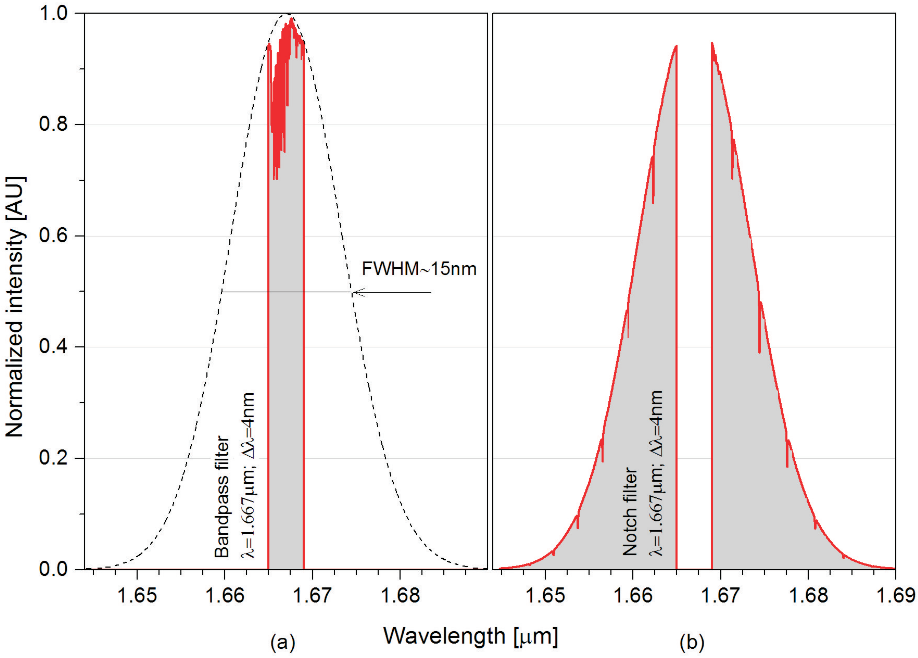

Fig. 2. Laser mode of 15 nm linewidth modulated by methane of 10 ppm GMR on 1.5 km lidar path: (a) λon signal transmitted by a bandpass filter of 4 nm linewidth; (b) λoff reference signal formed through a notch filter of matching linewidth

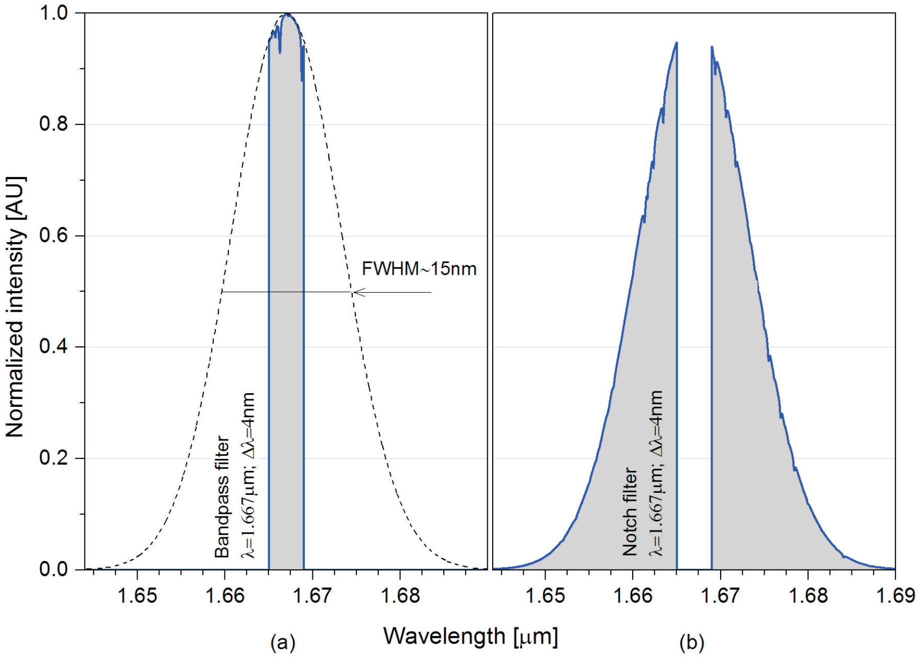

Fig. 3. Same laser mode of 15 nm linewidth modulated by water vapour alone of 10 gm^{-3} GMR on 1.5 km lidar path: (a) λon signal transmitted by a bandpass filter of 4 nm linewidth; (b) λoff reference signal formed through a notch filter of matching linewidth

mode. The laser line modulated by the joint resonance spectra of methane and water vapour is passed through a bandpass filter of 4 nm linewidth centred at 1.667 μm forming λon-signal (Fig. 2, 3). That spectral window is blocked in the shaded λoff-part with ratio of the laser pulsed energy per spectral channel set inversely proportional to the relevant spectral intensities of the segmented laser line of 0.25:0.75. The multiplexation of the lidar returns is performed with high efficiency, while the absorption by water vapour is equalized in both channels and eliminated in the DIAL signal. Custom spectral filters of the necessary parameters are planned to be ordered from manufacturers of similar commercially available products [11].

The absorption functions for calibration of the DIAL signal depicted in Fig. 4 correspond to the two types of laser diodes referenced in the Introduction. The graphical dependence based on an algorithm using Exp. 1 – Exp. 3 with the updated HITRAN'12 database and conventional analytical software Origin'8 is linear for the case of weak absorption by methane. The comparative parameter of pulse energy of 1 μJ per spectral channel is determined for rated pulse duration of the relevant laser diode. However, the laser diodes emitting longer pulses (Fig. 4a) result in undesired lower resolution. In addition, the revealed dependence on

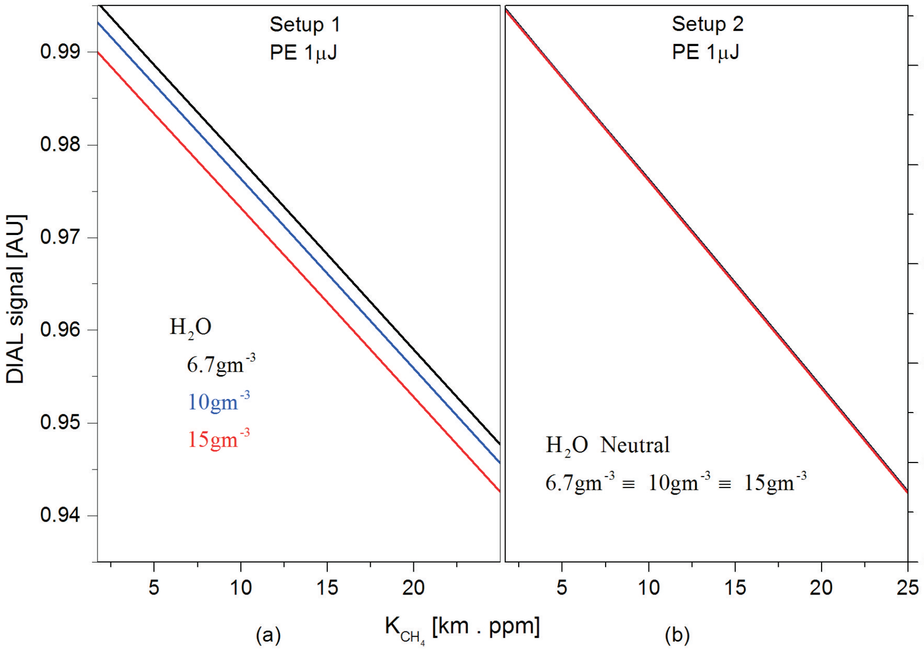

Fig. 4. Calibration functions vs. product K of gas mixing ratio and lidar path for different values of humidity and selected laser diodes of 1 µJ energy: (a) paired design of 1.625 µm/1.667 µm wavelengths, 8 nm linewidth and 10 µs pulse duration; (b) optimal design with multiplexation of radiation of 1.667 µm wavelength, 15 nm linewidth and 150 ns pulse duration

humidity degrades the detection limit. Conversely, the coincidence of the lines (Fig. 4b) verifies the invariance of measurement on humidity in perfect agreement with the theoretical result (Exp. 6). The functional dependence in Fig. 4b is also advantageous for detection of low gas mixing ratios of methane because of the twice-larger magnitude of the integral absorption.

Conclusions. A broadband lidar based on high-power laser diodes for remote sensing of the greenhouse gas methane is discussed in the study. An original differential absorption method is developed through multiplexation of the lidar returns modulated by the absorption spectra of methane and water vapour in dual spectral channels of the advanced optical scheme. The optimal detection of methane content unaffected by atmospheric humidity is verified on the basis of high-resolution spectral data, balancing the absorption of water vapour in the segmented broad laser line confined around 1.667 µm wavelength. Such an effective, compact and low energy-consuming DIAL system is prospective for wider deployment in the ecological monitoring, particularly, for mobile and airborne reconnaissance including energy resources and safety control of gas pipelines.

REFERENCES

[1] IPCC 2007 Summary for Policymakers Climate Change (2007) In: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change (Eds S. Solomon et al.), UK and NY USA, Cambridge University Press, 1–18.

[2] Miller S. M., M. A. Taylor, J. D. Watts (2018) Understanding high-latitude methane in a warming climate, Earth & Space Science News, **99**, 15–19.

[3] Penchev S., V. Pencheva, S. Naboko (2012) Atmospheric remote sensing hygrometer employing powerful broad-line laser diodes, C. R. Acad. Bulg. Sci., **65**(5), 669–674.

[4] Thomas B., G. David, C. Anselmo, J. P. Cariou, A. Miffre et al. (2013) Remote sensing of atmospheric gases with optical correlation spectroscopy and lidar: first experimental results on water vapor profile measurements, Applied Physics B, **113**, 265– 275.

[5] Thomas B., G. David, C. Anselmo, E. Coillet, K. Rieth et al. (2013) Remote sensing of methane with broadband laser and optical correlation spectroscopy on the Q-branch of the $2\nu_3$ band, Journal of Molecular Spectroscopy, **291**, 3–8.

[6] Platt U. (1994) Differential optical absorption spectroscopy (DOAS). In: Air monitoring by spectroscopic techniques (Ed. M. W. Zigrist), New York, John Wiley & Sons, 27–84.

[7] Riris H., K. Numata, S. Li, S. Wu, A. Ramanathan et al. (2012) Airborne measurements of atmospheric methane column abundance using a pulsed integrated-path differential absorption lidar, Applied Optics, **51**(34), 8296–8305.

[8] HITRANonline, HITRANonline data. Available at: `https://hitran.org/data-index/` [Accessed September 30, 2022].

[9] Laserdiode.com.2022. High Power Pulsed | OSI Laser Diode Inc. Available at: `https://www.laserdiode.com/standard_products/` [Accessed May 14, 2022].

[10] Penchev S., V. Pencheva, T. Dreischuh BG Utility model, Reg. No. 4239, 2022.

[11] Alluxa.com.2022. Ultra Narrow Spectral Filters. Available at: `https://www.alluxa.com/optical-filters/ultra-series-optical-filters-and-coatings/bandpass-filters/ultra-narrow-bandpass-filters/` [Accessed May 14, 2022].

Acad. Emil Djakov Institute of Electronics
Bulgarian Academy of Sciences
72 Tsarigradsko Shosse Blvd
1784 Sofia, Bulgaria
e-mail: `spenchev@ie.bas.bg`
`vasilka@ie.bas.bg`
`tanjad@ie.bas.bg`