MONITORING OF ATMOSPHERIC METHANE EMPLOYING DIAL SCHEME ON POWERFUL PULSED LASER DIODES

Stoyan Penchev, Vasilka Pencheva, Luben Mihov

Abstract. DIAL method is proposed for determination of the abundance of atmospheric methane. The spectral properties of laser radiation of high-power pulsed laser diodes are optimised within the molecular absorption bands of 1.55-1.625 μm of this major greenhouse gas. The characteristic broad laser line of the selected laser diodes modulated by the selected explicit absorption spectrum is explored by computational convolution based on HITRAN reference data on spectral linestrengths. The lidar scheme is ultimately compact, of low-energy consumption and suggests a large potential for ecological monitoring.

Keywords: Ecological monitoring, atmospheric methane, powerful laser diodes

1. INTRODUCTION

Atmospheric methane is a powerful greenhouse gas that plays a key role in several atmospheric environmental domains directly affecting the present enhancement of the greenhouse effect. It is the cause of about 20% of the increased trapping of atmospheric infrared radiation during the past 200 years. In addition, it affects the oxidizing capacity of the atmosphere and, therefore, the lifetimes of other strong greenhouse gases. The methane could be measured over regions of peak concentrations like sewage treatment and rice paddies [1]. While future technologies may limit methane emissions from human activities, the measurement of emissions from natural sources could be utilized - e.g., for prospecting of petroleum deposits [2].

Among the various techniques of detection of atmospheric methane, lidar remote sensing has the advantage of obtaining three-dimensional profiles with high temporal and spatial resolution unattainable by in-situ analysers. The proposed method optimises the spectral properties of radiation of selected powerful laser diodes within NIR molecular absorption bands of methane. The absorption spectrum is explored by a developed computational convolution method.

2. DIFFERENTIAL ABSORPTION OF LD RADIATION

The operation principle of DIAL (differential absorption lidar) scheme employs switching from an absorption line to a nearby wavelength with negligible absorption that makes use of both scattering and absorption processes. The DIAL method presented in this paper utilizes commercial high-power pulsed laser diodes by a combination of a series of the spectral characteristics of radiation. We have proposed DIAL dual-beam set-up, which employs 100 mW pulses tuned on, and off the intensive methane rovibrational absorption spectrum confined within 1.55-1.625 μm, undisturbed by overlapping atmospheric major gases water vapour and carbon dioxide. The resultant wavelength shift is feasible for high-resolution DIAL measurement of negligible systematic error due to the nonresonant spectral properties of the atmosphere (Rayleigh and Mie scattering). A computational convolution method is developed to utilise the potential of the selected laser diodes for spectroscopic and DIAL applications. The mathematical consideration of the resonance absorption of the broad laser line is based on Beer-Lambert's law [3], assuming that the profile of a separate absorption line is not resolved (Fig.1). The resultant integral form of the absorption spectrum is derived as a function of frequency [4-7] modulated by the laser profile in the measured tuning range.

The resonance absorption of laser intensity is evaluated assuming spectral linestrengths modulated by a laser line of Gaussian profile and absorption lines of Lorentzian profile. The spectrum contains the absorption coefficients of over 50 lines of methane molecule imported from HITRAN spectral database [http://hitran.jao.ru/]. The ratio of initial and absorbed laser radiation is derived according to Beer-Lambert’s law. The investigated spectral range is processed by resolved step of 0.1 cm⁻¹:

\[ A = \int \left| \frac{I}{I_0} \right| dv = \int \exp \left\{ -\frac{(v - v_i)^2}{\Delta v_i} - \sum_{n} \frac{f_n(v, v_n, v_o)}{L} \right\} dv \quad (1) \]

where \( I, I_0 \) stand for the intensity of initial and absorbed laser radiation; \( \Delta v_i \) is width of Gaussian laser line; summation is by the evaluated and
experimentally corrected absorption coefficients $k_n$ [cm$^{-1}$]; $v_n$ and $\Delta v_n$ are centre frequencies and width of absorption lines and coefficient $L$ [cm] is lidar (double-pass) path.

![Fig.1. High-resolution rotation- vibration absorption spectrum of methane molecule broadened by atmospheric pressure containing separate lines convolved by Gaussian laser line centred at 6124 cm$^{-1}$ stabilised frequency; by heating to 40$^\circ$C](image)

The explicit DIAL signal is determined summing all absorption lines and integrating them numerically in the relevant modulation range. The resultant atmospheric integral absorption of laser radiation on lidar path is by a convolution integral, taking into account in a second approximation the dependence of the complex profile of the laser line on pulse duration:

The validation of DIAL measurement is based on equation (1) and HITRAN linestrength database calibrated experimentally with reference to a high-resolution spectrometer.

3. SENSOR OF BIOGAS METHANE BASED ON LD COUPLE

The integral absorption spectrum of biogas methane constrained in the DIAL signal is found spectrally pure in the range around 1.6$\mu$m, whereas the coefficients of maximal absorption is unsaturated providing a great dynamic range of measurement. Laser-diodes of InGaAsP compound [8] of wavelength matching the strong absorption band of methane around 1.63 $\mu$m are particularly advantageous. This type refers to highly reliable devices for a range of applications in severe environmental conditions with operating temperatures up to 70 $^\circ$C. They generally exhibit broadened multimode laser line of typically 8nm spectral width owing to the large emitting surface. The proposed rovibrational spectrum of methane is accessible by a feasible system employing the selected laser diodes in a single chip coupled with optical fibre, which also matches the standard broadband interference filters for suppression of ambient sunlight.
Fig.2. High-power InGaAsP laser diodes of broad-line pulsed laser radiation matching the methane molecular absorption spectrum of 6124 cm\(^{-1}\) frequency [http://www.laserdiode.com]

<table>
<thead>
<tr>
<th>Table 1. Typical parameters of MCW563S series</th>
</tr>
</thead>
<tbody>
<tr>
<td>Port 1</td>
</tr>
<tr>
<td>--------</td>
</tr>
<tr>
<td>Power</td>
</tr>
<tr>
<td>Centre wavelength</td>
</tr>
<tr>
<td>Spectral width</td>
</tr>
<tr>
<td>Threshold current</td>
</tr>
<tr>
<td>Test conditions</td>
</tr>
</tbody>
</table>

Except the structural feasibility, the integral character of the detected spectrum simplifies the critical operating regimes of thermal stabilization of the lasers and frequency tuning between the different lines characteristic of other DIAL systems.

Methane profound absorption spectrum at 1.63 \(\mu\)m is situated outside the major absorption of water vapour and carbon dioxide (Fig.3). The relevant combination of lasers allows measurement of systematic error less than 1% set by the spectral dependence of the nonresonant Rayleigh and Mie scattering. The estimated detection limit on 100m track is equivalent to resolution of 100 ppm of the urban atmospheric abundance, which is competitive to other sensors based on differential absorption [see, e.g., 9]. The selected spectrum is referenced by the theoretical values on Fig.3.

The integral absorption spectrum is practically immune of variations of atmospheric pressure which is an important advantage. This factor with reference to other DIAL techniques based on detection of a single absorption line of the gas molecule may result in considerable measurement error though the system possesses the necessary detection limit.

**CONCLUSIONS**

Biogas methane is a powerful greenhouse gas with strong absorption bands around 1,5-1,65 \(\mu\)m wavelengths appropriate for implementation of the high-power laser diodes of InGaAsP technology. The prospect of development of DIAL application for monitoring of the atmospheric abundance of methane and other atmospheric greenhouse gases like water vapour and carbon dioxide is set on the
parameters of their integral absorption spectra matching the characteristic lower spectral resolution of the radiation of these lasers. Such system is ultimately compact, of low-energy consumption and suggests a potential for ecological monitoring which could supplement or replace the available instruments.

Fig. 3. Evaluated atmospheric transmission vs. frequency of laser radiation within the selected feasible tuning ranges of high-power pulsed laser diodes

REFERENCES


7. S. Penchev, V. Pencheva, V. Naboko, S. Naboko (Application of Pulsed GaAs Diode Lasers to High


МОНИТОРИНГ НА АТМОСФЕРНИЯ МЕТАН С ИЗПОЛЗВАНЕ НА ДИАЛ СХЕМА НА МОЩНИ ИМПУЛСНИ ЛАЗЕРНИ ДИОДИ

С. Пенчев, В. Пенчева, Л. Михов

Резюме. Предложен е ДИАЛ метод за определяне на концентрацията на атмосферния метан. Спектралните свойства на лазерното лъчение на мощни импулсни лазерни диоди са оптимизирани в молекулярния абсорбционен спектър 1.55-1.625 mm на този основен парников газ. Характерната широка линия на лазерните диоди модулирана от избрана абсорбционен спектър е изследвана чрез компютърна конволюция на основата на силата на резонансните линии спектрална в база данни HITRAN. Лидарната схема е изключително компактна, с ниска енергийна консумация и се предполага да има голям потенциал за екологичен мониторинг.

Keywords: Ecological monitoring, atmospheric methane, powerful laser diodes

Stoyan Penchev
Institute of Electronics, Bulgarian Academy of Sciences
blv.Tzarigradsko shousse, 72
Sofia 1784
E-mail: spenchev@ie.bas.bg

Vasilka Pencheva
Institute of Electronics, Bulgarian Academy of Sciences
blv.Tzarigradsko shousse, 72
Sofia 1784
E-mail: vasilka@ie.bas.bg

Luben Mihov
Institute of Electronics, Bulgarian Academy of Sciences
blv.Tzarigradsko shousse, 72
Sofia 1784
E-mail: mihovli@abv.bg