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IMPROVING LASER METHANE EMISSION MONITORING WITH FREQUENCY–AMPLITUDE FEATURE OF ABSORPTION PULSE

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Annotation. Accurate monitoring of methane emissions is vital for environmental management and safety in oil and natural gas operations. This paper introduces a methodology leveraging advanced analytics and experimental data to build a linear regression model for methane concentration estimation. The approach utilizes a time–frequency analysis and incorporates proposed frequency–amplitude features of the absorption pulse, which have shown a strong correlation with methane concentration. Experimental results validate that the proposed feature achieves a higher correlation (R^2=0.981) with methane concentration.

Keywords. Laser absorption spectroscopy, time–frequency, methane emission, absorption pulse, frequency–amplitude.

Introduction. Methane is a colorless, odorless and flammable gas that can be found in a variety of environments, including natural gas production and distribution facilities, landfills, agricultural operations and industrial plants. Detecting methane leaks is important for safety, environmental and regulatory reasons. Optical methods for determining methane gas concentration employ spectroscopic techniques, providing accurate and reliable measurements. Infrared absorption spectroscopy is a prominent approach within this category, where methane molecules absorb specific wavelengths of infrared light. The concentration of methane is based on the level of light absorption, offering a highly sensitive and selective detection method. There exist three major approaches based on tunable diode laser absorption spectroscopy: direct absorption spectroscopy (DAS), wavelength modulation spectroscopy (WMS) and wavelength modulation–direct absorption spectroscopy (WM–DAS).

DAS is the most straightforward version of laser absorption spectroscopy due to limited equipment required for its implementation, quick alignment and interpretation of raw data. [1, 2]. However, this method has lower sensitivity due to uncertainty in the laser output signal measurement. In contrast to DAS, WMS using the harmonic detection technology can effectively eliminate the influence of baseline uncertainty and work with a much higher modulation frequency (as high as several hundred kHz) [3, 4] . Therefore, WMS is resistant to low–frequency noise and has a higher sensitivity. As it is known, WMS technology has been widely studied and used for gas properties measurement [5, 6, 7, 8]. While WMS can achieve higher accuracy and sensitivity in gas absorption measurements compared to DAS, the complexity of signal modulation and the use of a lock–in amplifier make WMS more challenging to implement than the DAS method. WM–DAS combines the simplicity and calibration–free nature with the enhanced noise rejection and high sensitivity offered by the harmonic detections, establishing it as an efficient and reliable method for methane gas concentration estimation.

This paper implements advanced analytics techniques on experimental datasets to build a linear regression model for estimating methane concentration. A larger experimental dataset improves model accuracy by capturing complex relationships between independent variables and the dependent variable. Additionally, larger sample sizes are more representative of the overall population, thereby reducing variance–related errors.

In our paper, we focus on analyzing the raw absorption signal features related to methane gas concentration obtained by laser absorption spectroscopy in time–frequency domain. The main contributions of our research are summarized as follows:

– The proposed frequency–amplitude feature of absorption pulse is proposed to improve methane concentration estimation accuracy;

– The proposed linear regression model for estimating methane concentration;

– A detailed step–by–step methodology for estimating the features of the absorption pulse is presented, enhancing the precision and reliability of the estimation process.

The method for methane absorption pulse features estimation. The transmitted light intensity $I_t(t)$ of the radiation can be related to the absorbing gas concentration and presented by the Beer–Lambert law, is given by:

$$I_t(t) = I_0(t) \cdot exp[-\sigma(\lambda)cl] \tag{1}$$

where $I_0(t)$ is the laser output signal; λ is the wavelength of laser emitter; $\sigma(\lambda)$ is the absorption coefficient, which is a function of wavelength λ; *c* is the gas concentration; l is the optical path length.

When performing the harmonic analysis [9], the detected signal $I_t(t)$ in Eq. (1) can be divided as:

$$I_t(t) = \left\{(V_{DC} \propto \bar{I}) + \left(V_{indep_{1f}} \propto i\right) + \left[\left(V_{dep_{2f}} + V_{dep_{3f}} + V_{dep_{4f}} + \cdots\right) \propto (\sigma cl)\right]\right\} \tag{2}$$

where V_{DC} represents the DC signal depending on the average intensity $\bar{I}$; $V_{indep_{1f}}$ is the absorption independent $1f$ component which is related to the intensity scanning depth i; $V_{dep_{2f}}, V_{dep_{3f}}, V_{dep_{4f}}, \ldots$ are amplitudes of harmonics.

As follow from Eq. (1) and (2) transmitted intensity signal $I_t(t)$ consists two main components: laser output and absorption signals. Based on the literature review, it can be confirmed that the method for methane absorption pulse features estimation (**Figure 1**) is based on procedures including signal decomposition [9], signal calibration [5], segmentation and fusion of pulse in absorption signal (as show in Absorption pulse extraction session).

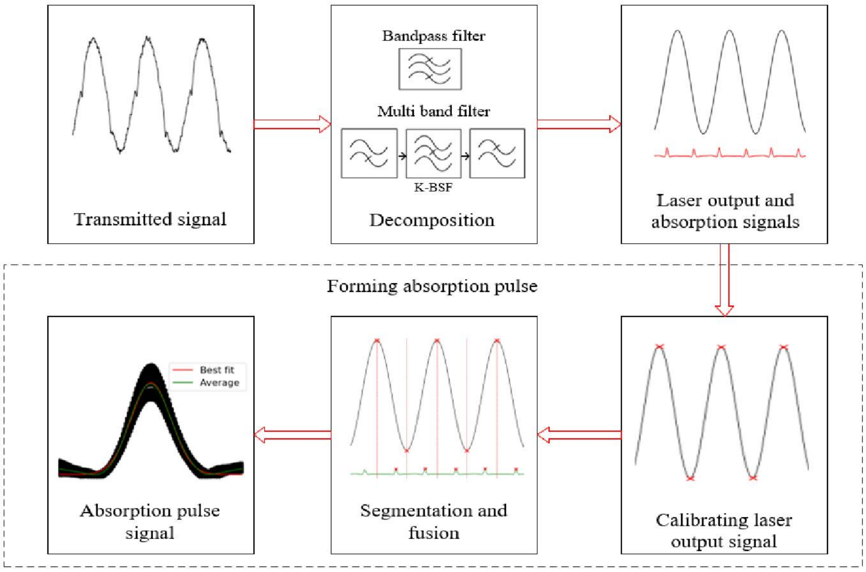

Figure 1 – The method for methane absorption pulse features estimation

In the final step, the absorption pulse signal $AP_{I_{ATSMA}}$ is defined in the time domain. For estimating the absorption pulse features related to gas concentration, it is proposed the following two frequency–amplitude features of absorption pulse.

1.The amplitude spectrum of absorption pulse.

$$A_{F_{AP}} = \max_{1 \leq k \leq N_F} |F_{AP}(k)| \quad (3)$$

where $|F_{AP}(k)|$ is the k–th spectral sample value of the spectrum of absorption pulse $AP_{I_{ATSMA}}$; N_F is the number of spectral samples of the spectrum of absorption pulse.

2.The integrated spectrum of absorption pulse.

$$F_{APint} = \sum_{k=1}^{N_F} F_{AP}(k) \quad (4)$$

Laser output and absorption signals extraction. The aim of this process is to extract the laser output and absorption signals. **Figure 2** shows the transmitted signal.

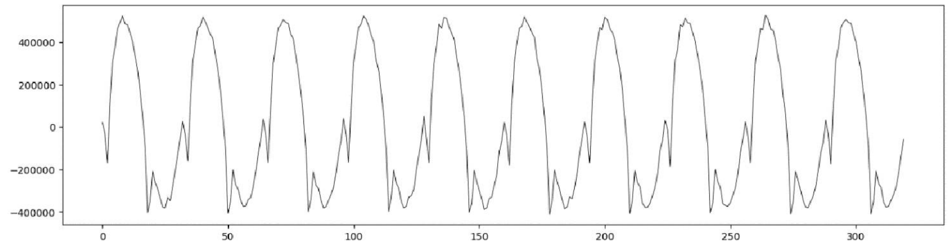

Figure 2 – Transmitted signal

After analyzing the transmitted signal, the laser output signal is extracted by applying bandpass filter with center frequency 10 kHz and bandwidth 100Hz, as shown in **Figure 3**. This signal represents the initial laser light and serves as a reference for calibrating absorption signal.

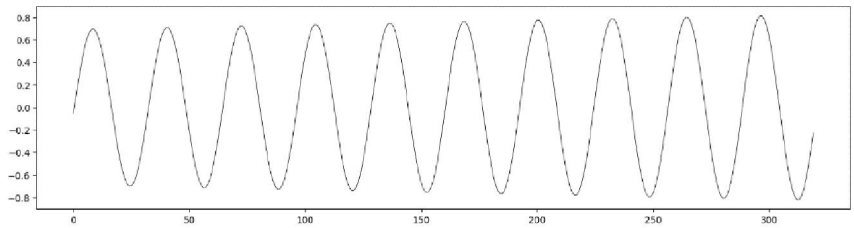

Figure 3 – Laser output signal

The absorption signal is extracted by Fourier–domain–based line shape recovery method [9. and represented in **Figure 4**. This signal is critical for determining the concentration of methane, as it represents the amount of laser light absorbed by the methane molecules.

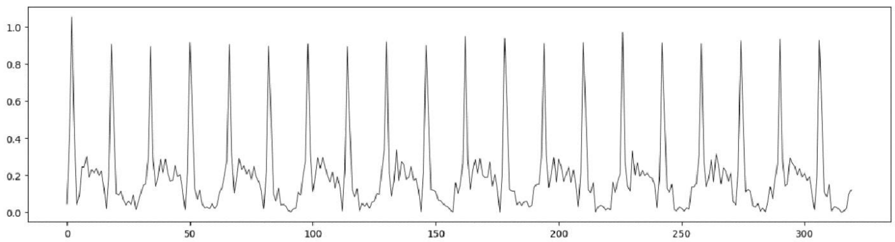

Figure 4 – Absorption signal

Absorption pulse extraction. In this section, the focus is on the extraction of the absorption pulse from the laser output and absorption signals. To begin the extraction process, the absorption signal is segmented (red dot line) based on peak and valley of the laser output, as depicted in **Figure 5**. This segmentation helps in isolating the pulse interval of the absorption signal that correspond to each half cycle of the laser output signal.

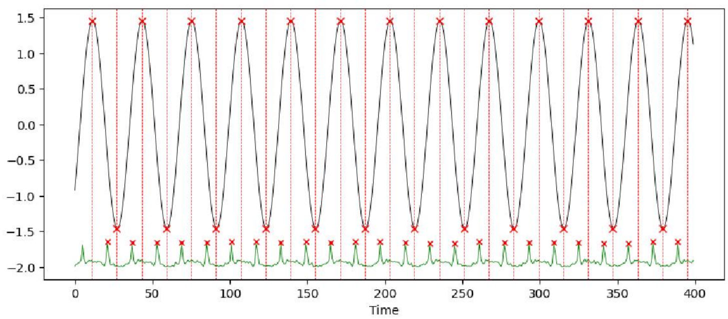

Figure 5 – Illustration of the segmentation of the absorption signal

As the laser intensity transitions from a local maximum to a local minimum, the laser's wavelength sweeps across the methane absorption line. The absorption line in spectroscopy refers to a specific wavelength where a target gas molecule absorbs laser light and it is less affected by other gases. When the laser wavelength coincides with the methane absorption line, the absorption intensity reaches its peak.

Once the segmentation is completed, the next step involves averaging all the segmented absorption pulses. **Figure 6** shows the averaged absorption pulse and its best fit curve.

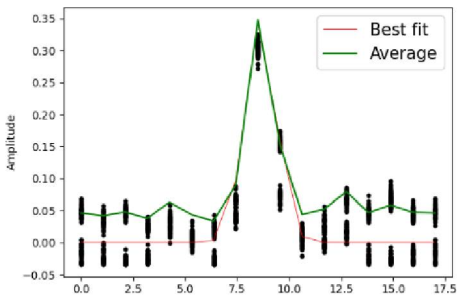

Figure 6 – The averaged absorption pulse signal

These steps are essential for accurately extracting and analyzing the absorption pulse, which is a key factor in absorption pulse features estimation.

Absorption pulse features estimation in frequency domain. Analyzing the absorption pulse in the frequency domain allows for the extraction of key features that characterize the methane absorption process, which can be more effective than time–domain analysis. The process begins by applying a Fourier transform to the averaged absorption pulse, converting the signal from the time domain to the frequency domain. **Figure 7** presents the frequency spectrum of the absorption pulse.

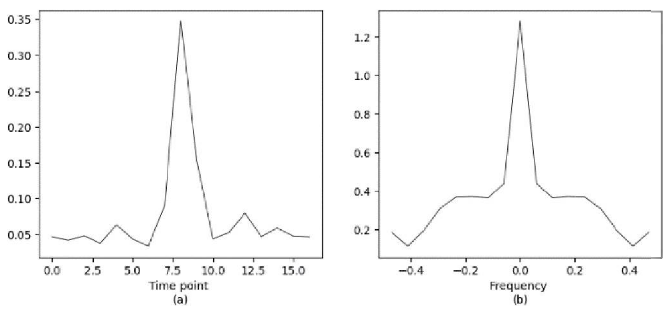

Figure 7 – Absorption pulse in time (a) and frequency domain (b)

The features of absorption pulse in frequency domain provide valuable insights into the methane concentration and can be used to enhance the accuracy of concentration measurements.

Estimation of the relationship between CH_4 concentration and absorption pulse features. The experiment was performed with laser output power of 166nW passing through a methane volume with different concentrations:100 ppm, 250 ppm, 540 ppm, 1000 ppm and 2000 ppm. For each of five concentration value, 5 measured signals will be used.

Relationship between methane concentration and well–known absorption pulse features, such as absorption pulse amplitude [10] and integrated absorption pulse [11] in time domain, was compared with proposed frequency–amplitude features of absorption pulse (amplitude spectrum and integrated spectrum of absorption pulse) as shown in **Figure 8**. The coefficient of determination R^2 is used as a measure of goodness of fit.

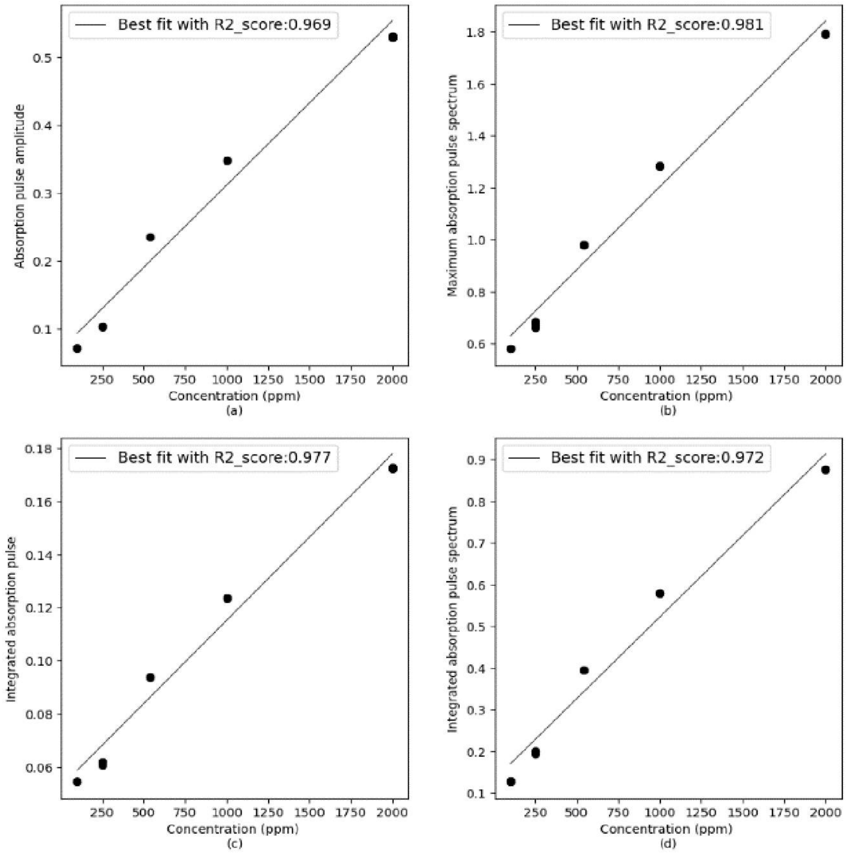

Figure 8 – Relationship between the absorption pulse features and methane concentration: (a) absorption pulse amplitude (R2=0.969), (b) amplitude spectrum of absorption pulse (R2=0.981), (c) integrated absorption pulse (R2=0.977), (d) integrated spectrum of absorption pulse (R2=0.972)

It follows from **Figure 8** that the linear regression model $y = 0.00064x + 0.566$ with $R^2 = 0.981$ (**Figure 8 (b)**), where x is CH_4 concentration value and y is amplitude spectrum of absorption pulse, is more robust. It is mentioned that with a large amount of experimental data, the linear regression model can achieve higher accuracy.

Conclusion. The two frequency absorption pulse features (amplitude spectrum and integrated spectrum **of absorption** pulse) related to gas concentration have been proposed. These features were derived from a time–frequency analysis of the absorption signal, and they significantly enhance the correlation of methane concentration and absorption signal feature. These features show a strong correlation with methane concentration, with an $R^2 = 0.981$, indicating their effectiveness in accurately estimating methane concentrations.

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