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TIME DELAY-PHASE HARMONIC MODEL OF TRANSMITTED LASER SIGNAL FOR GAS CONCENTRATION ESTIMATION

Bach N.V.

Belarusian State University of Informatics and Radioelectronics, Minsk, Republic of Belarus

Tsviatkou V.Yu. – Doctor of Technical Sciences, Professor

Annotation. The proposed time delay-phase harmonic model takes into account absorption, scattering, and reflectance effects and the relationship between the phase shift and the time delay and the absorption pulse signal quality. The Levenberg–Marquardt algorithm is applied to define optimal model parameters, enabling an efficient and effective of gas concentration estimation process. Experimental results demonstrate a strong correlation between the harmonic ratio feature and methane concentration under varying laser power conditions. Furthermore, the phase shift effect on the absorption signal is investigated, revealing its impact on waveform symmetry and peak distribution. The proposed method has been proven to require only a few signal cycles for gas concentration estimation.

Keywords. Tunable diode laser absorption spectroscopy, gas concentration, Levenberg–Marquardt algorithm, harmonic model, time delay, phase shift.

Introduction. One of the most promising approaches for remote methane detection, even at extremely low concentrations, is optical methods. Tunable diode laser absorption spectroscopy (TDLAS) is among the most effective optical techniques for this purpose [1, 2]. There are three primary variations of TDLAS: direct absorption spectroscopy (DAS) [3, 4], wavelength modulation spectroscopy (WMS) [5, 6], and wavelength modulation-direct absorption spectroscopy (WM-DAS) [7, 8].

Direct absorption spectroscopy is the simplest form of TDLAS, requiring minimal equipment and offering straightforward alignment and data interpretation. However, its sensitivity is limited due to inherent uncertainty in the laser output signal measurements. On the other hand, wavelength modulation spectroscopy (WMS) overcomes this limitation by using harmonic detection technology, which enables higher modulation frequencies-sometimes reaching several hundred kHz-thereby improving sensitivity and eliminating baseline uncertainty. As a result, WMS is more resilient to low-frequency noise and offers enhanced measurement accuracy. This technology has been widely studied and implemented for gas property measurements.

Although WMS provides higher accuracy and sensitivity compared to direct absorption spectroscopy, it is more complex to implement due to the need for signal modulation and the use of lock-in amplifiers. The WM-DAS method combines the simplicity and calibration-free benefits of direct absorption spectroscopy with the high sensitivity and noise rejection capabilities of harmonic detection, making it a powerful and reliable technique for methane concentration estimation.

The goal of our paper is to develop harmonic models of the interaction laser light with gas for providing the trade-off between computational complexity, robustness and accuracy of gas concentration estimation. The key contributions of our paper can be summarized as follows:

- the proposed harmonic model of the absorption pulse signal is based on time delay and phase shift and optimal model parameter estimation influencing on the signal pulse quality related to gas concentration;
- the harmonic ratio feature of an extracted-and-formed absorption pulse defined by the ration of the sum of the even harmonic amplitudes of absorption pulse signal to the fundamental harmonic amplitude is proposed for estimating gas concentration.

Methodology of the harmonic model integration. Based on the analysis of the harmonic properties of the absorption pulse signal $I_{abs}(t,\lambda)$, the proposed time delay-phase harmonic model of this signal is defined as:

$$I_{abs}(t,\lambda,\Delta t,\varphi) = \sum_{k=1}^{K_H/2}\left(C_{2k}(\lambda)\sin(2\pi\cdot 2kf_1\cdot(t+\Delta t)+\varphi)\right), \tag{1}$$

where $C_{2k}=\sqrt{A_{2k}^2+B_{2k}^2}$ is the amplitude of the *k*-th even harmonics, Δt and φ are the time delay and phase shift of the absorption pulse signal influencing the position and shape of absorption pulses.

The absorption pulse signal $I_{abs}(t,\lambda,\Delta t,\varphi)$, fundamental harmonic signal $I_{RAW}^{f_1}(t)$, DC component $I_{DC}(t)$ and the attenuation coefficient K_{env} are computed by the Levenberg–Marquardt algorithm [9]. The Levenberg–Marquardt algorithm provides a trade-off between computational complexity and concentration accuracy.

Given a set of *m* empirical samples $\left(t, I_{tr}(t,\lambda,K_{env})\right)$ of independent and dependent variables, we find the optimal parameters β_{opt} from $\beta=\{A_1,B_1,I_{DC},K_{env}\}\cup\{C_{2k}\}_{k=1,\dots,K_H}$ by using the objective function represented as:

$$\beta_{opt} = \underset{\beta}{argmin} \sum_{t=1}^{m} \left(I_{tr}(t, \lambda, K_{env}) - \hat{I}_{tr}(t, \lambda, \beta) \right)^2, \quad (2)$$

where $\hat{I}_{tr}(t,\lambda,\beta) = \left(I_{DC}(t,\lambda) + I_{RAW}^{f_1}(t,\lambda) + I_{abs}(t,\lambda,\Delta t,\varphi) \right) \cdot K_{env}(\lambda)$ is the estimated transmitted laser intensity signal.

Values $\Delta t = \left(arctan\left(\frac{B_{2k}}{A_{2k}}\right) - \varphi \right) / (2\pi \cdot 2kf_1)$ is defined from φ and $\{A_{2k}, B_{2k}\}$, which are computed by the objective function (2).

The harmonic ratio feature of absorption pulse A_{ab} is defined as the sum of the harmonic component's amplitudes of absorption pulse signal $I_{abs}(t,\lambda,\Delta t,\varphi)$ divided by the amplitude of the fundamental component $I_{RAW}^{f_1}(t,\lambda)$ for reducing systematic noise, represented as:

$$A_{ab}(\lambda) = \frac{\sum_{k=1}^{K_H/2} C_{2k}(\lambda)}{C_1(\lambda)}, \quad (3)$$

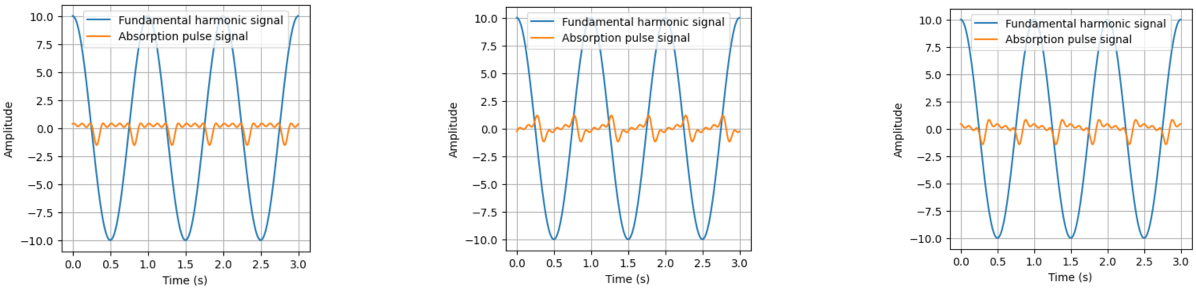
where $C_1 = \sqrt{A_1^2 + B_1^2}$ is the amplitude of the fundamental harmonic.

Estimation of the linear weight coefficients of the even harmonics. The Levenberg–Marquardt algorithm based on using the objective function (2) allows us to estimate the linear weight coefficients A_{2k} and B_{2k} (Table 1) for the most significant even harmonics $K_H = 7$.

Table 1 – Estimation of the linear weight coefficients (A and B) of the even harmonics under laser power 166 nW

Even harmonic indexes *k*	Methane gas concentration									
	100 ppm		250 ppm		540 ppm		1000 ppm		2000 ppm	
	A	B	A	B	A	B	A	B	A	B
2	-12580.3	-15060.3	-15949.8	-18362.2	-41067.3	-44922.1	-57938.1	-61931.8	-84645.2	-88710.2
4	-21423.6	493.9	-26671.6	784.3	-55775.1	2751.2	-76085.5	4227.8	-98583.9	5551.1
6	-4972.6	6505.5	-9722.10	12307.5	-19997.9	26414.4	-32743.2	42695.6	-48527.9	63852.8
8	1119.6	2610.1	1934.44	7812.8	4542.6	24875.2	7049.8	36113.5	11825.8	61218.7
10	3961.8	2352.2	6853.37	3801.7	12326.7	7339.7	22683.9	13926.6	37705.4	23071.7
12	4812.1	-1420.9	6224.06	-1600.9	13857.6	-3812.7	21908.2	-5800.3	33656.4	-9528.7
14	1140.3	-2185.3	2210.94	-5131.0	3209.2	-6334.2	5966.1	-11647.9	9882.9	-19649.9

Estimation of the phase shift value of absorption signal. Figure 1 shows influence of the phase shift values ($-\frac{\pi}{2}$, $-\pi$, $-\frac{\pi}{4}$) on the shape, amplitude and symmetry of the absorption pulses of signal $I_{abs}(t,\lambda,\Delta t,\varphi)$ using time delay-phase harmonic model.



a) b) c)

Figure 5 – Influence of the phase shift values on the absorption signal pulse parameter:
a) $\varphi = -\frac{\pi}{2}$, b) $\varphi = -\pi$, c) $\varphi = -\frac{\pi}{4}$

If $\varphi = -\frac{\pi}{2}$, then the positive part of the absorption pulse signal tends to flatten, while the negative part reaches two extrema within one fundamental period. If $\varphi < -\frac{\pi}{2}$, then the positive component skews higher on the right. If $\varphi > -\frac{\pi}{2}$, then the positive component shifts higher on the left, and the negative part does not reach its minimum value as in the case of $\varphi = -\frac{\pi}{2}$.

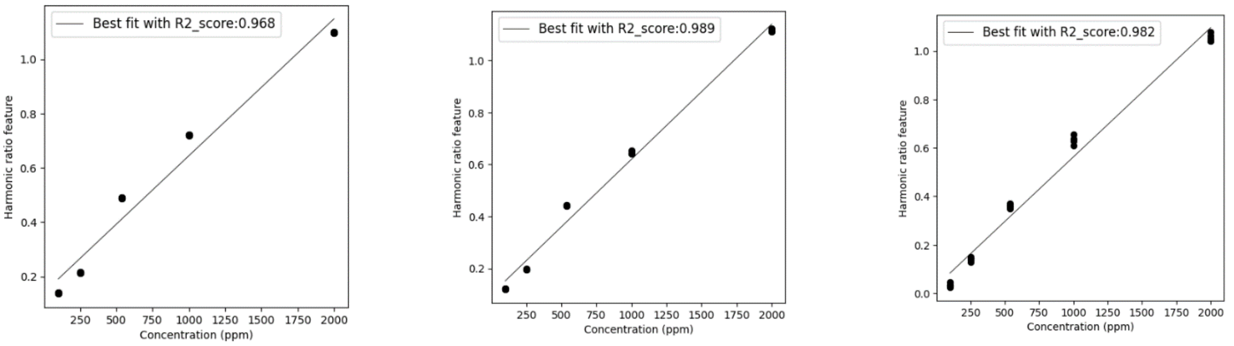
The optimal phase shift value of the absorption signal is determined to be $\varphi = -\frac{\pi}{2}$ due to the laser light absorption of the target gas.

Estimation of time delay value of the absorption pulse signal. After defining parameters A_{2k}, B_{2k} and φ, the parameter Δt is computed for the laser power of 166 nW (Table 2). It follows from Table 2 that the time delay of the second harmonic is equal to -5.58×10^{-5} s, while for the other harmonics, it is approximately equal to -6.60×10^{-6} s.

Estimation of gas concentration based on the time delay-phase harmonic model. Linear regression models of the relationship between the harmonic ratio feature A_{ab} and methane concentration under the time delay of the second harmonic −5.58×10^{-5} s, and other harmonics -6.60×10^{-6} s, $\varphi = -\frac{\pi}{2}$ for the most significant even harmonics $K_H = 7$ are shown in Figure 2. Using the proposed time delay-phase harmonic model (1) and the objective function (2), we find the optimal parameters β_{opt} and then the harmonic ratio feature of absorption pulse A_{ab} corresponding to concentration under different laser powers.

Table 2 – Estimation of the time delay of the absorption pulse signal under a laser power of 166 nW

Time delay Δt (s) (×10^{-6})	Even harmonic index *k*	Methane gas concentration				
		100 ppm	250 ppm	540 ppm	1000 ppm	2000 ppm
-55.8	2	-55.5	-55.7	-55.9	-56.0	-56.1
-6.60	4	-6.34	-6.37	-6.45	-6.47	-6.47
-6.60	6	-6.60	-6.56	-6.61	-6.60	-6.61
-6.60	8	-7.06	-6.73	-6.61	-6.63	-6.63
-6.60	10	-6.65	-6.69	-6.65	-6.62	-6.63
-6.60	12	-6.63	-6.58	-6.61	-6.59	-6.62
-6.60	14	-6.60	-6.68	-6.61	-6.60	-6.61



a) b) c)

Figure 2 – Relationship between the harmonic ratio feature A_{ab} and methane concentration under different laser powers: a) 166 nW, b) 9 nW, and c) 2 nW

It follows from Figure 2 that the correlation coefficient R^2 under different laser powers is close to 1, which indicates a good fit between our observed and expected data. The time delay and phase shift values are not affected by variations in laser power. It is determined that the absorption signal energy is concentrated in the lower even harmonics.

Conclusion. The proposed method optimizes gas absorption parameters by an objective function used in the Levenberg–Marquardt algorithm. Experimental results demonstrate a strong correlation ($R^2 > 0.97$) between the harmonic ratio feature and methane concentration across different laser power changes, validating the robustness of the method. Additionally, the impact of phase shift on waveform symmetry and peak distribution is analyzed, highlighting its significance in signal interpretation. The time delay for the second harmonic is equal to −5.58×10^{-5} s, for higher harmonics it is approximately −6.60×10^{-6} s and a phase shift of $\varphi = -\frac{\pi}{2}$ allow us to reduce time complexity by decreasing the number of parameters in the proposed models.

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