DOI 10.31489/2023No4/111-115

UDC 53.043

COHERENT AMPLITUDE MODULATION IN A LASER GAS SENSOR

Zhanabaev Z.Zh., Akhtanov S.N., Tileu A.O., Almen D.B.*

Al-Farabi Kazakh National University, Almaty, Kazakhstan, dinara.almen@gmail.com

The article is devoted to a current topic in science and technology - determining the type and concentration of gas using a laser. At close values of green laser radiation intensity and bias voltage, interference patterns of the time series of the photodiode output signal were obtained. The degree of coherence (more than ~0.1) made it possible to distinguish between the types of gases and their concentrations. Signal coherence was controlled by Allan deviation values. The novelty of the research method is the choice of the harmonic of the electrical network as the main modulation frequency. It is shown that the correlation functions and the corresponding power spectra are sensitive to low-frequency fluctuations of molecules and their clumps. This allows the results and methods of this work to be used in specific cases instead of large-sized and expensive complexes of optical instruments at room pressure and temperature.

Keywords: laser, gas sensor, low frequencies, photodiode, ammonia.

1. Introduction

Determining the presence of a target gas and monitoring its concentration is an important task in technology, medicine, safety, and environmental monitoring. Extensive research is devoted to the choice of element sensitivity, electrical, and optical circuits of the sensor in order to increase its efficiency. Laser absorption spectroscopy systems have been created and are used in more than 1000 coal mines in China [1,2]. In recent years [3,4], the laser absorption spectroscopy method has been significantly advanced by using a Herriott multiphase cell, which makes it possible to detect a target gas with a concentration of $\sim 10^2$ parts per million in total over an averaging time of $(1 - 10^2)$ s. The review [2] indicates a general pattern of signals from sensors on nanofilms with different chemical compositions – the predominance of low-frequency noise (LFN). In LFN spectroscopy, the intensity of the noise signal is usually inversely proportional to frequencies in the range $(10^2 - 10^5)$ Hz.

Note that LFN signals are also present in laser spectroscopy. Laser radiation passing through a gas is absorbed and scattered not only on individual atoms with a certain resonant frequency, but also on individual molecules and their clumps. As a result, a fluctuation radiation field is created. At the output of the photodiode, this field interacts with the bias potential to create a partially coherent interference pattern. The correlators or power spectra of low-frequency fluctuations depend more noticeably on the gas concentration than on its atomic structure. An important task of gas sensors is to determine maximum allowable concentrations (MACs) leading to explosion, poisoning, and technological violations. Critical gas concentrations are usually determined by chemical analysis. Physical processes that determine the MACs can be nonlinear conductivity of the photodiode, thickening, and noisy spectral lines. Perhaps for this reason, there is insufficient data in the literature on the results for concentrations ppm $\geq 10^3$ obtained by physical methods.

From the listed factors of weakening the sensor response with increasing gas concentration, it follows that it is necessary to select an intense low-frequency oscillation in the form of amplitude modulation.

In terms of the general principles of amplitude modulation to identify the desired signal in our approach, the following roles are meant: carrier frequency - laser frequency, fundamental modulation frequency - harmonics from the power supply network of electrical devices, the influence of the concentration of the target gas realizes intermodulation distortion (heterodyning). In this case, the result will separate the effect of changing the electrical resistance of the nanoelements of the sensor from the concentration effect.

The purpose of the work is to experimentally obtain the time-fluctuating voltage at the output of a photodiode as the laser passes through a gas layer. Under conditions of the required degree of coherence,

obtaining correlation spectral patterns. Search for opportunities to establish maximum permissible gas concentration values without the use of large, expensive optical analysis systems.

2. Measuring principle

The experimental setup diagram is shown in Figure 1. A TL081CP converter was used to amplify the output time-fluctuating voltage $v(t)$from PIN. The distance between the laser source and the photodiode is 5-6 cm. At photodiode offset voltage $U = -2.3$ V (in the region of stability of the PIN characteristic) and photon energy $\hbar\omega$ =2.3 eV, the beam intensity is $IU = 60$ mA*2.3 V = 0.138 J.

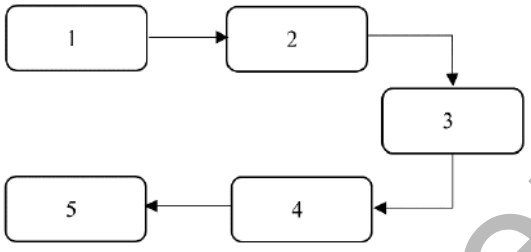


Fig.1. Laser gas sensor assemblies: 1 – power supply unit for the laser source "GOPHERT GPS-3205", 2 – source of a laser beam with a wavelength of λ=532 nm, 3 – vessel (volume 20 ml) of gas for an air+gas mixture, 4 – PIN photodiode, 5 – measuring instruments current voltage NI ELVIS II+, Hantex.

Various interference patterns were obtained $v(\hbar\omega, -U, I)$, degree of coherence determined by $(v_{min}/v_{max})^2$, $\gamma \geq 0{,}1$. The intensity of the laser beam has a pronounced maximum current value $I\sim$ 60 mA. All measurements were carried out at normal atmospheric pressure and room temperature.

3. Results and Discussion

Figure 2 shows voltage fluctuations $v(t)$ obtained when a laser beam with a wavelength of λ=532 nm passes through air, ammonia, and ethanol. The gas concentration is $\lesssim 10^3$ ppm. Value intervals $v(t) =$ 1-1.35 V, observation time t =120 s. The results were obtained at photodiode supply voltage $U =$2.3 V, laser beam photon energy in units of electron-volt $\hbar\omega =$2.3 eV. Due to the interaction of low-frequency fluctuations caused in the medium by the laser and harmonics of instrumental sources, interference patterns are observed.

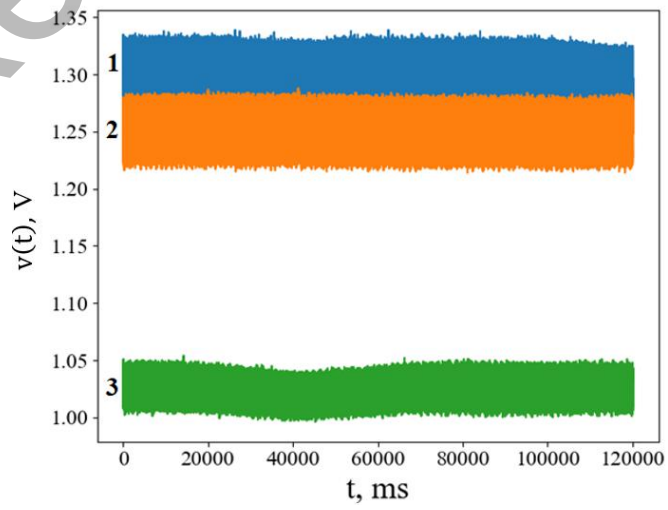


Fig.2. Voltage fluctuations at the photodiode output: 1-air, 2-ammonia, 3-ethanol.

The degree of coherence of interference fringes is determined by the formula:

$$\gamma = \frac{[1 - (\mathrm{v_{min}}/\mathrm{v_{max}})^2]}{[1 + (\mathrm{v_{min}}/\mathrm{v_{max}})^2]} \quad (1)$$

where $\mathrm{v_{min}}$, $\mathrm{v_{max}}$ are the minimum and maximum values of the fluctuation voltage $\mathrm{v(t)}$. Formula (1) uses normalized intensity values.

For the analysis, results with $\gamma \geq 0{,}1$ were used, which corresponds to the recommendations accepted in optics for choosing the contrast of interference patterns. Fluctuations are stationary, but nonequilibrium. The average values $\langle \mathrm{v(t)} \rangle$ are shifted: for air (low beam absorption), the maximum for the rest is lower. A universal characteristic of complex processes is correlation functions that consider the time delay of the count. For fluctuation processes, it is customary to call this function abbreviated as correlators, which are determined from discrete reports of fluctuations ($\delta v_i, \delta v_j$):

$$K = < \delta v_i, \delta v_j > = \frac{1}{N^2} \sum_i^N \sum_j^N \delta v_i * \delta v_j \quad (2)$$

Correlators can distinguish quantitatively different gases relative to air at a fixed concentration. Figure 3 shows autocorrelation functions depending on time t and its displacement τ.

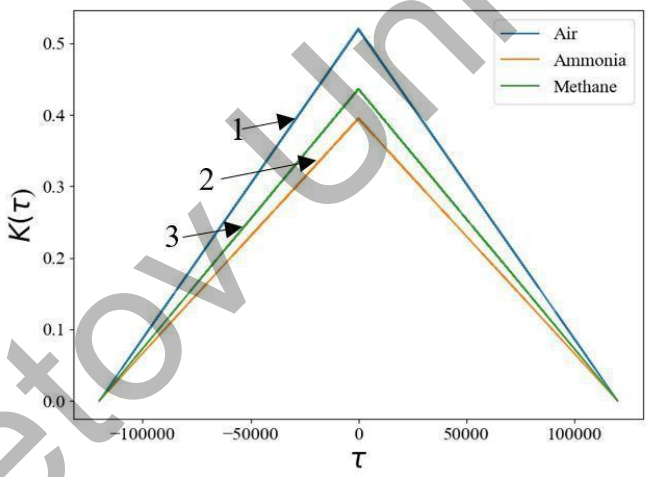


Fig.3. Autocorrelation function of fluctuation v(t):1-air, 2-ammonia, 3-methane.

A change in the laser sensor signal depending on the concentration $\mathrm{(C)}$ of the target gas is also observed in the power spectrum. Let us introduce the notation $\mathrm{K_{g/a}}$– the ratio of the correlators of gas (g) and air (a). Then the expression $\mathrm{ln} < \delta v_i, \delta v_j >_{g/a}$ describes the difference $\mathrm{K_g}$ from $\mathrm{K_a}$ more accurately than their direct difference $\mathrm{K_g - K_a}$. Figure 4 shows the power spectra obtained from the correlators at the observed intense harmonic with a frequency of $f = 200$ Hz. As the concentration of $\mathrm{NH_3}$ increases, the intensity of the lines decreases. At $\mathrm{C} \geq 10^3$ppm (parts per million), this trend is ambiguous.

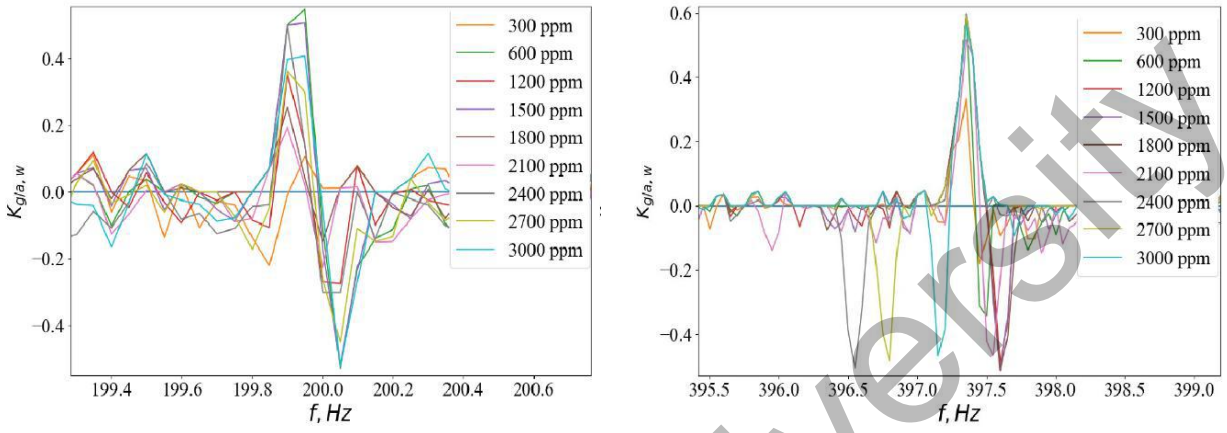
As noted, with increasing irradiation intensity and gas concentration current, the signal from the sensor can become saturated in a nonlinear manner. In this case, the role of an individual factor is unknown in advance. The given example at fixed frequencies clearly separates the effect of concentration.

The proposed approach to coherent amplitude modulation sensing is a variation of various radio engineering, solid-state methods. Low-frequency noise, kilohertz, megahertz ranges are used; there are names of wave modulation, frequency modulation, and nanoparticle spectroscopy [5-8].

In recent years [8-10], Allan deviation has been widely used in gas sensors, which is found by considering the average values of fluctuations of successively divided windows of time samples. In our

approach, the standard deviation ($\sigma > \sigma_{\min}$) has a stable value (Figure 5) due to coherence, while in the above works it increases after a minimum.

Different stable values of σ correspond to different concentrations. The σ_{min} values were determined by repeated measurements. This result indicates an average accuracy of 0.5% of the present experiment on the fluctuation signals. The measurement accuracy of survey work is ±0.5 Hz (2.5%) at frequency. From Figure 4 the same order of deviation from the fundamental frequency of various additional modulations follows.



a b

Fig.4. Ammonia power spectra: a) Main modulation frequency $f_0 = 200$ Hz; b) Main modulation frequency $f_0 = 397$ Hz.

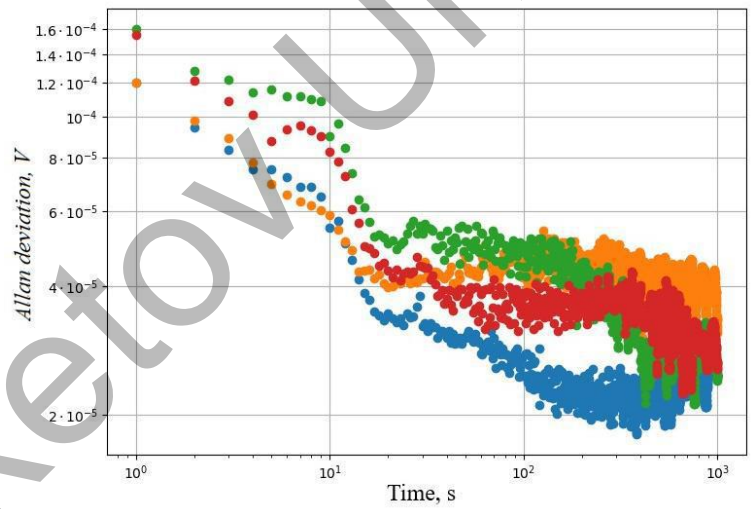


Fig.5. Allan deviation for NH_3 (ppm value: $10^2 \div 10^3$).

4. Conclusion

The possibility of determining the type and concentration of gas in the air with a laser sensor by analyzing low-frequency fluctuation signals was experimentally demonstrated. The difference from existing optical sensors is the use of an interference pattern of interaction between harmonics of measuring instruments and fluctuation modes of absorption and laser scattering. From the results of the experiment, conclusions were drawn for the first time about the possibility of using a method for describing and predicting maximum permissible gas concentrations without knowledge of atomic and molecular spectral lines. The proposed laser gas sensor technology opens the possibility of creating a software package, since harmonic frequencies, spectrum expansion parameters (diffusion coefficient, sizes of Brownian nanoparticles, their mean free path) are known experimentally and there are tables for various gases. These parameters are determined much more accurately in comparison with quantum optical and dispersion

characteristics, since at high optical frequencies these characteristics will be nonlinear and ambiguous. The work was performed using the simplest equipment. The main result is the detection of saturation of the gas sensor response with increasing gas concentration through low-frequency modulation of the fluctuation signal. The work may find various applications by improving, if necessary, experimental techniques.

Acknowledgments

We would like to express our sincerest gratitude to the Al-Farabi Kazakh National University for supporting this work by providing computing resources (Department of Physics and Technology).
This research was funded by the Committee of the Ministry of Science and Higher Education of the Republic of Kazakhstan, grant AP14872061.

References

1 Gong W., Hu, J., Wang Z., Wei Y., Li Y., Zhang T., Zhang Q., Liu T., Ning Y., Zhang W., Grattan K.T.V. Recent advances in laser gas sensors for applications to safety monitoring in intelligent coal mines. *Frontiers in Physics*, 2022, Vol.10, pp. 1058475. doi: 10.3389/fphy.2022.1058475

2 Shin, Wonjun Hong, Seongbin Jeong, Yujeong Jung, Gyuweon Park, Jinwoo Kim, Donghee Choi, Kangwook Shin, Hunhee Koo, Ryun-Han Kim, Jae-Joon Lee, Jong-Ho. Low-frequency noise in gas sensors: A review. *Sensors and Actuators B: Chemical*, 2023, Vol. 383, pp. 133551. doi: 10.1016/j.snb.2023.133551.

3 Liu, Kun Wang, Lei Tan, Tu Guishi, Wang Zhang, Weijun Chen, Weidong Gao, Xiaoming. Highly sensitive detection of methane by near-infrared laser absorption spectroscopy using a compact dense-pattern multipass cell. *Sensors and Actuators B: Chemical*, 2015, Vol.220, pp. 1000 – 1005.

4 Zhang Z., Li, M., Guo J., Du B., Zheng, R. A Portable Tunable Diode Laser Absorption Spectroscopy System for Dissolved CO2 Detection Using a High-Efficiency Headspace Equilibrator. *Sensors*, 2021, Vol.21, pp. 1723. doi:10.3390/s21051723

5 Hua-Liang Yu, Jun Wang, Biao Zheng, Bing-Wen Zhang, Li-Qin Liu, Ying-Wu Zhou, Cheng Zhang, Xiao-Ling Xue. Fabrication of single crystalline WO 3 nano-belts based photoelectric gas sensor for detection of high concentration ethanol gas at room temperature. *Sensors and Actuators A: Physical*, 2020, Vol.303, pp.111865. doi:10.1016/j.sna.2020.111865

6 Liu C., Zheng L., Wang J., et al. Tunable terahertz gas laser based on a germanium spectrum splitter. *Appl. Phys. B*, 2020, Vol.126, pp. 133. doi:10.1007/s00340-020-07486-5

7 Bierer B., Grgić D., Yurchenko O., Engel L., Pernau H.-F., Jägle M., Reindl L., Wöllenstein J. Low-power sensor node for the detection of methane and propane. *J. Sens. Sens. Syst*., 2021, Vol. 10, pp. 185–191. doi:10.5194/jsss-10-185-2021.

8 Wang Y., Feng Y., Adamu A.I., Dasa M.K., Antonio-Lopez J.E., Amezcua-correa R., Markos C. Mid-infrared photoacoustic gas monitoring driven by a gas-filled hollow-core fiber laser. *Scientific reports*, 2021, Vol. 11, No.1, pp. 3512.

9 Kwak D., Lei Y., Maric, R. Ammonia gas sensors: A comprehensive review. *Talanta*, 2019, Vol. 204, pp.713 – 730. doi: 10.1016/j.talanta.2019.06.034.

10 Dudzik G, Abramski K. Solid-state laser intra-cavity photothermal gas sensor. *Sensors and Actuators B: Chemical*, 2021, Vol. 328, pp. 129072.

Article received 04.12.2023
Article accepted 18.12.2023