Photoacoustic Application for Gas Detection

Mohammed A. Hussain
Department of Laser and Optoelectronics Engineering, Al-Nahrain University, Iraq

Israa Amir Kadhim
Department of Laser and Optoelectronics Engineering, Al-Nahrain University, Iraq
missisraa89@yahoo.com

Abstract

Laser photo acoustic spectroscopy is a technique to detect and measure trace gas using infrared laser radiation. This technology is based on the generation of acoustic wave in a gas excited by a modulated laser beam at a wavelength corresponding to the absorption line of the gas species at the resonance frequency of the acoustic resonator, and detecting this sound wave using a sensitive microphone.

This work presents the experimental procedure of laser photoacoustic gas detection at different levels of concentration using Freon-12 as the sample gas.

The experimental work has been accomplished by fabricating a photoacoustic cell which consists of an acoustic resonator of 50mm length and 4mm diameter. The resonator is terminated by two gas buffers and partial transmission windows for laser wavelength of 10.6µm. A sensitive microphone has been fixed in midpoint between the mirrors. A CO_2 laser of effective power levels 0.7-2W has been used to induce the acoustic wave.

The analysis of experimental data has been accomplished using matlab software.

Keywords: laser Photoacoustic spectroscopy, acoustic harmonics, and gas detection.

1. Introduction

The photo acoustic (PA) effect is based on the sample heating produced by optical absorption of modulated radiation [1, 2]. Periodic heating and cooling of the sample is necessary to generate pressure fluctuations of the gas which generates acoustic waves that can be detected by sensitive sound detectors such as microphones or piezoelectric transducers [3].

A photo acoustic spectroscopy (PAS) for gas detection is shown in figure 1. Gas molecules are irradiated by intermittent infrared wave at resonance frequency. The gas molecules absorb some of the incident light energy and excited from the ground state to an excited state. Some of the excited molecules are transferred into translational motion by means of nonradiative relaxation process, then the gas temperature is raised and the gas pressure increases. When the incident light is modulated at a given frequency, gas temperature increase is periodic at the modulation frequency, so it causes a cyclical change of pressure. The sound wave is picked up by microphone and is converted into electric signal which is then tested and analyzed by external circuits. The creation and detection processes of photoacoustic signal are a transformation process of light, heat, sound and electricity [4].

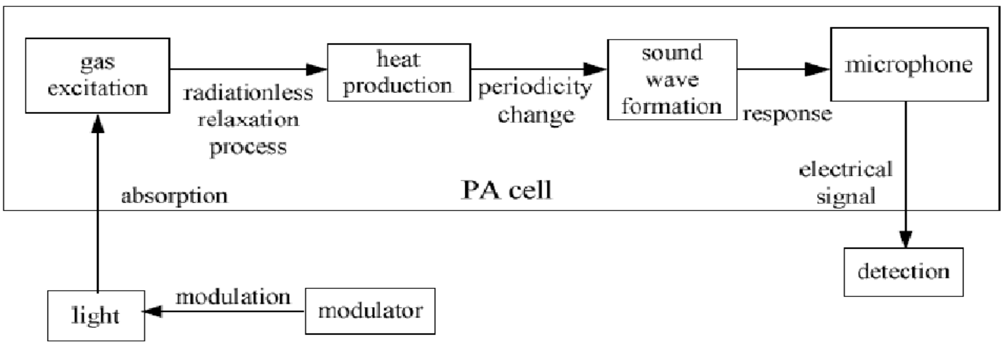

Figure 1: Photo acoustic spectrometry diagram for gas detection [4].

PAS is recognized as a zero background technique (i.e. in the absence of any absorbing gas, there is no acoustic signal) [1, 5, 6]. This offers several advantages including wide dynamic range, nondestructive detection, simplicity and compact size. Based on the advantages mentioned above, it has been widely used in various fields such as atmospheric sciences [7,8] and industrial process control [9, 10]. Laser PAS method can also be used for the detection of a wide variety of industrial gases including benzene, hydrogen cyanide, acetylene, carbon monoxide, carbon dioxide, and poisonous gases such as hydrogen cyanide, explosives (TNT and PETN)[11and12], combustion process and pollution monitoring [7 and8]. PAS techniques have also been used in medical diagnosis that promise to revolutionize the manner in which diagnostics are carried out today and may soon lead to rapid diagnostics in terms of improved results, decreased cost, and expanded life span[11].

Photo acoustic spectroscopy is a technique that has high selectivity to distinguish the gas species present in a multicomponent gas mixture such as air, and high the sensitivity is essential to detect a very low concentrations of substances.

Photo acoustic spectroscopy has consolidated one of the most sensitive and versatile in the monitoring of gases. Detection limits in the bands of part per billion by volume (ppbV, 10^{-9}) and part per trillion by volume (pptV, 10^{-12}) [9].

2. Experimental procedure

Photoacoustic experiments were performed using an arrangement (figure 2) which consists of the following: CO_2 laser, home-made acoustic resonator, miniature microphone, F-12 gas canister, a personal computer and gas supply accessories.

A CO_2 laser of 10.6μm wavelength modulated at different frequencies ranging from 750Hz to 7500Hz with two effective power levels of 0.7 and 2W to induce photoacoustic waves inside the resonator in the presence of Freon-12.

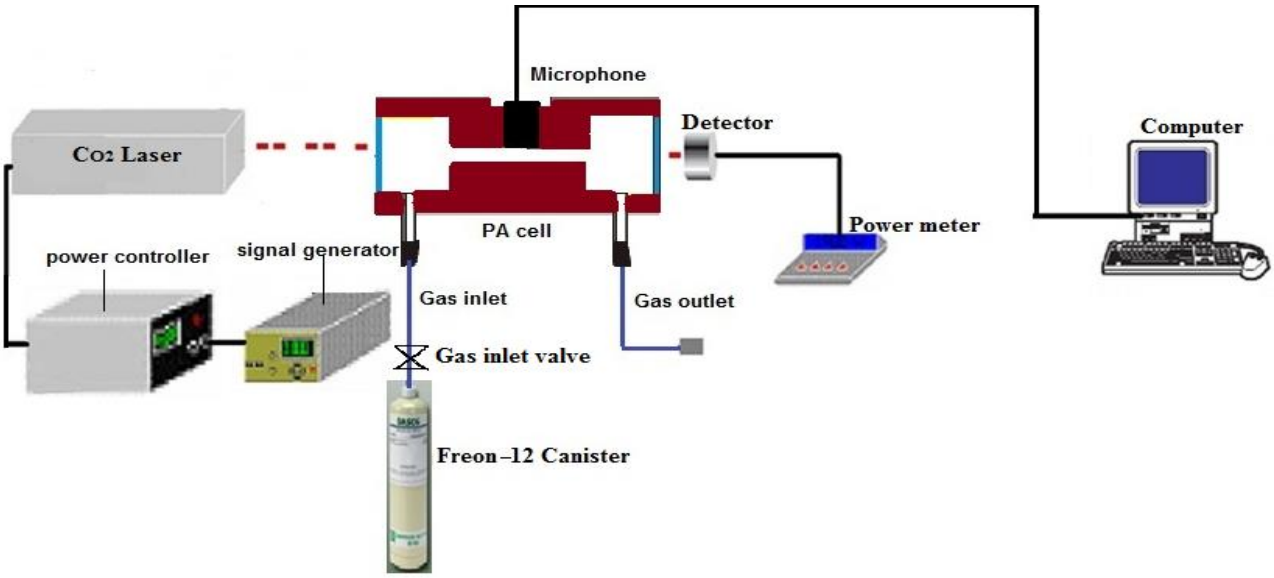

Figure 2: Schematic picture of the experimental arrangement.

The modulated laser beam is passed through the PA cell through ZnSe mirror, while the generated pressure waves are detected using a sensitive microphone as acoustical signal. The latter is displayed on the screen of a computer using software-sound card oscilloscope.

The first measurement of the acoustic wave was recorded while Freon-12 is flowing across the resonator bore to assure positive filling with the gas, i.e.100% gas concentration inside the resonator. Lower gas concentrations are achieved by closing the gas inlet valve while the outlet valve is open to induce natural gas exchange with atmospheric air.

Different laser modulation frequencies, higher and lower than the calculated resonance frequency were forced into the PA cell to study the gas response accordingly.

The resonance frequency of Freon-12 has been calculated for the present resonator to be 1500Hz and that of atmospheric air is 3462Hz at normal conditions according to Eq.(1) [2,13] and Eq.(2) [2,14] below, using Freon & air properties at normal conditions:

$$f_{kmn} = \frac{v_s}{2}[(\frac{k}{L})^2 + (\frac{\alpha_{mn}}{\pi r})^2]^{\frac{1}{2}} \quad \text{..... (1)}$$

Where v_s sound velocity in the sample gas, L and r are the length and radius of the resonator; k, m and n indices (non-negative integers) refer to the values of the longitudinal, azimuthal, and radial modes, respectively, and α_{mn} is the n_{th} root of the derivative of the m_{th} Bessel function.

$$v_s = (\frac{\gamma RT}{M})^{\frac{1}{2}} \quad \text{..... (2)}$$

Where γ the ratio of the specific heat of the gas at constant pressure C_p to that at constant volume C_v, R is the ideal gas constant, T is the absolute temperature, and M is the molecular weight.

PA signals and their frequencies spectrum have been recorded for different modulation frequencies which are higher and lower than the resonance frequency of Freon-12.

3. Experimental Results

Acoustic signal and their corresponding spectrums have been recorded and analyzed for different laser modulation frequencies and as follows:

a) Results at 1500Hz modulation

A pure acoustic signal without noise at 1500Hz (figure 3a) when the gas fills the cell has been observed. This is due to the good match between applied modulation frequency and the resonance frequency of Freon-12. Figure 3b shows frequency spectrum of the acoustic signal that illustrates the energy distribution of peaks frequencies. It is evident that most of the energy is stored in the fundamental harmonic, while the flat positive and negative peaks indicate wave saturation.

Figure 4 shows acoustic signal with harmonics at 1500Hz and 10s after closing the gas inlet valve, the Freon-12 concentration is reduced by mixing with air inside the acoustic cell. The 1500Hz modulation frequency does not fully match the resonance frequency of this mixture, which causes the 9th harmonic of the resonance frequency of Freon to appear as a second peak at 13500Hz. The intensity of fundamental frequency has dropped sharply due to the heavy molecular weight of Freon which results in depressing Freon molecules to the bottom of the cell bore, i.e. away from the laser path, beside the fast replacement of Freon with air which reduced light absorption.

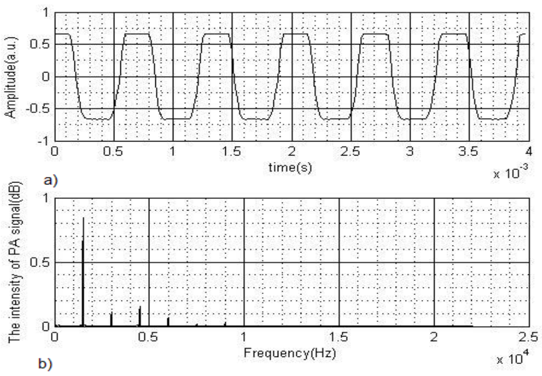

Figure 3: PA experimental results for Freon-12 at 1500Hz and full concentration as a) signal amplitude and b) frequency spectrum

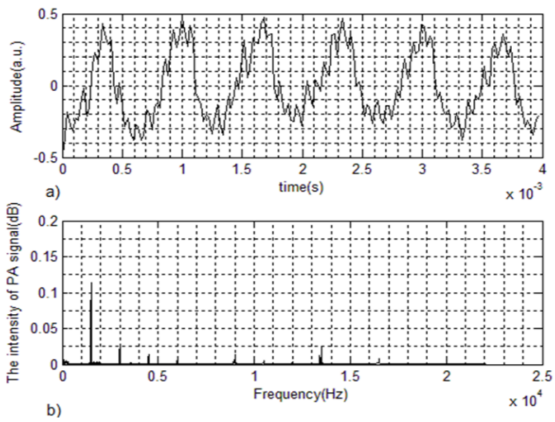

Figure 4: PA experiment results for Freon-12 at 1500Hz and 10s after closing inlet valve as a) signal amplitude and b) frequency spectrum

The intensity of the 2nd harmonic frequency at 3000Hz has grown after 15min from inlet valve closing as shown in figure 5 b. This indicates increased air concentration inside the PA cell while the 4th harmonic (6000Hz) has increased significantly and other harmonics appeared with minor intensity. However, the global energy storage in these harmonics is quite low due to reduced absorption of air molecules compared with Freon.

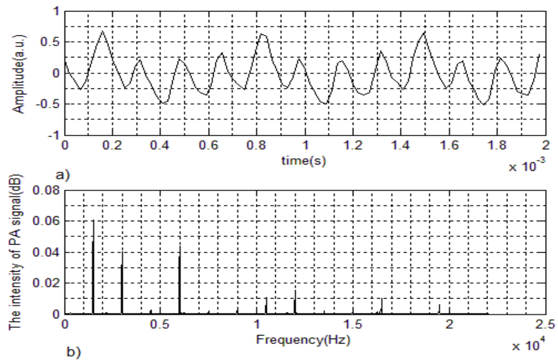

Figure 5: PA experiment results for Freon-12 at 1500Hz and 15minute after closing inlet valve as a) signal amplitude and b) frequency spectrum

b) Results at 750Hz modulation

Acoustic signal profile the second harmonic of 750Hz at full gas concentration is quite identical to that of 1500Hz as figure 6a reveals. The second harmonic of 1500Hz collects most of energy because this satisfies the resonance frequency of Freon, as can be inferred from figure 6b.

Many higher order harmonics (beside the fundamental) appeared after 5minute from inlet valve closing when the concentration of the air increased inside the cell, with the most significant being the 3rd,4th and 9th.

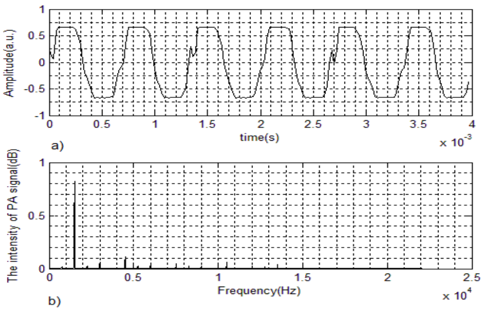

Figure 6: PA experimental results for Freon-12 at 750Hz and full concentration as a) signal amplitude and b) frequency spectrum

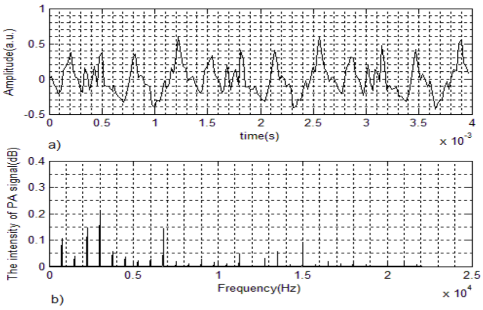

Figure 7: PA experiment results for Freon-12 at 750Hz and 5minute after closing inlet valve as a) signal amplitude and b) frequency spectrum

c) Results at 3500Hz modulation

Acoustic signal at 3500Hz modulation frequency and full gas concentration present many harmonics such as 2nd, 3rd and 4th as shown in figure 8. The strongest of which is the 4th (10500Hz). The same is repeated at 1minute after closing inlet valve (figure 9). This is because 10500Hz represents the 3rd harmonic of the resonance frequency for air, at the same time this is the 7th harmonic for Freon.

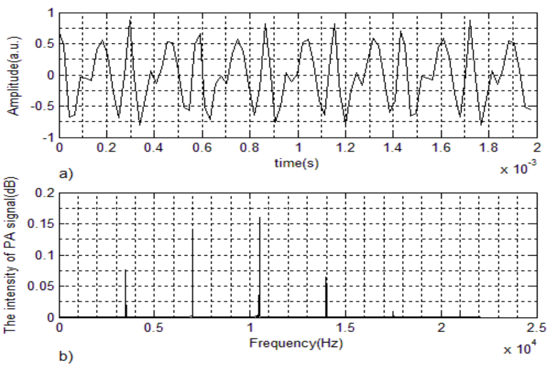

Figure 8: PA experimental results for Freon-12 at 3500Hz and full concentration as a) signal amplitude and b) frequency spectrum

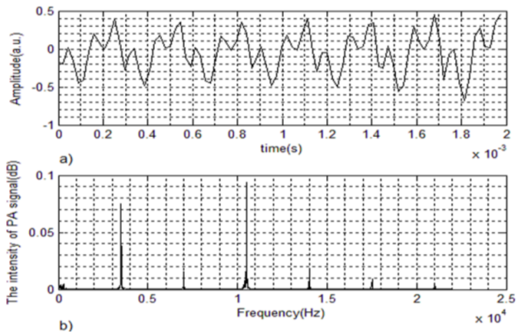

Figure 9: PA experiment results for Freon-12 at 3500Hz and 1minute after closing inlet valve as a) signal amplitude and b) frequency spectrum

d) Results at 6500Hz modulation

Frequency spectrum of acoustic signal at 6500Hz (figure 10) introduces a new phenomenon which is the sub-harmonics of modulation frequency of 2166Hz and 4332Hz. Though higher harmonics have been noticed, the highest is at 13000Hz which is the 2nd harmonic of 6500Hz and the nearest to the 9th harmonic of 1500Hz.

Figure11 shows that the strong peak at 2166Hz after 1minute from inlet valve closing. Higher harmonics are can also be seen.

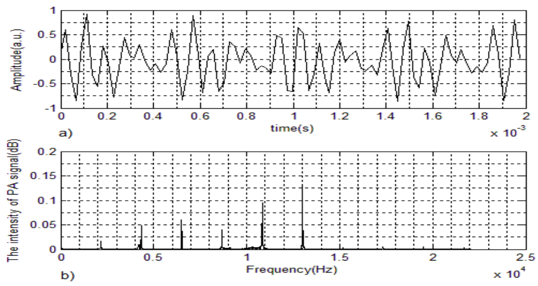

Figure 10: PA experimental results for Freon-12 at 6500Hz and full concentration as a) signal amplitude and b) frequency spectrum

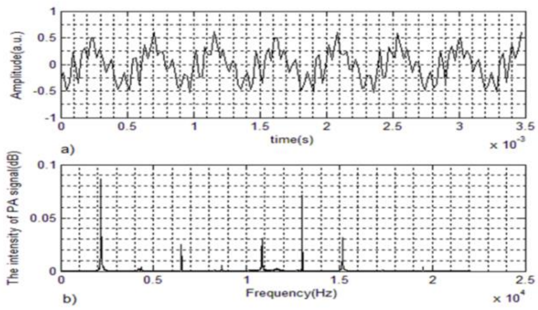

Figure 11: PA experiment results for Freon-12 at 6500Hz and 1minute after closing inlet valve as a) signal amplitude and b) frequency spectrum

The highest peak in figure 12 is located at 17400Hz after 5minute from inlet valve closing. This peak nearest to 3rd harmonic of air (19500Hz), at the same time it is nearest to 12th harmonic of Freon. Other harmonics of have appeared, yet at very low levels of energy.

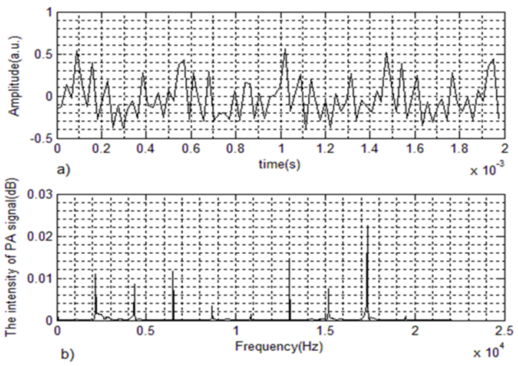

Figure 12: PA experiment results for Freon-12 at 6500Hz and 5minute after closing inlet valve as a) signal amplitude and b) frequency spectrum

e) Results at 7500Hz modulation

Figure 13 depicts the acoustic signal and frequency spectrum for 7500Hz laser modulation. Sub-harmonics have also been noticed at low energies. The strongest harmonic is sited at 10000Hz at full gas concentration which nearest to the 7th harmonic of the Freon.

Figure 14 shows low energy sub and higher harmonics at 1minute after closing inlet valve. These peaks are most probably representing the harmonics of the gas mixture in the cell. 15000Hz is the 2nd harmonic the modulation, representing the 10th harmonic of Freon and is nearest to 4th harmonic of the air.

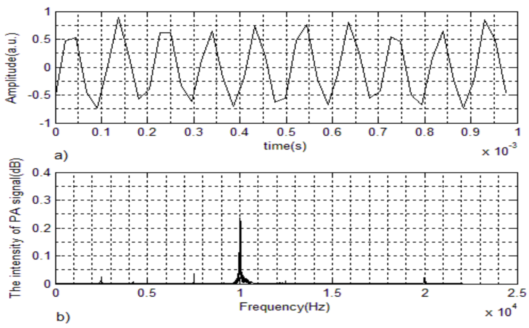

Figure 13: PA experimental results for Freon-12 at 7500Hz and full concentration as a) signal amplitude and b) frequency spectrum

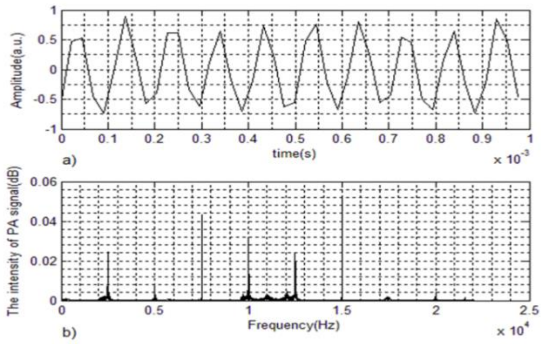

Figure 14: PA experiment results for Freon-12 at 7500Hz and 1minute after closing inlet valve as a) signal amplitude and b) frequency spectrum

Figure 15 shows a graph of the resonant frequency of the photoacoustic cell at full Freon gas concentration. This graph verifies that the frequency of 1500 Hz optimizes the operation of the photoacoustic cell, which is the calculated frequency for the operation of the acoustic cell filled with pure freon.

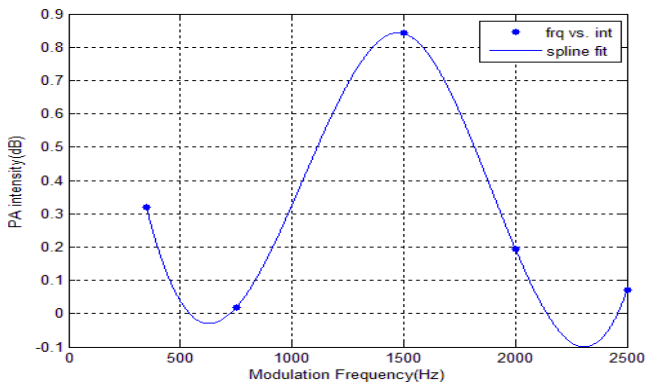

Figure 15: Variation of photoacoustic signal intensity with modulation frequency at full Freon concentration

4. Conclusions

The following conclusions have been deduced:

1. The gas concentration is interpreted according to line spectrum of the fundamental and higher harmonics or sub harmonics.
2. The optimum resonant frequency of the photoacoustic cell at full Freon gas concentration has been proved experimentally to be 1500Hz.
3. The threshold of modulation frequency for the present resonance cell is 6000Hz. Though it is possible to operate outside this threshold frequency with the possibility of inducing sub-harmonics of the modulation frequency.

References

[1] S. Schilt, F.K. Tittel and K.P. Petrov, "Diode Laser Spectroscopic Monitoring of Trace Gases", Encyclopedia of analytical chemistry, pp.1-29, 2011.
[2] D. C. Dumitras," CO_2 Laser optimization and application", InTech,Janeza Trdine 9, 51000 Rijeka, Croatia, ISBN 978-953-51-0351-6, 2012.
[3] D.V. Bageshwar, A.S. Pawar, V.V. Khanvilkar and V.J. Kadam," Photo acoustic Spectroscopy and Its Applications", Eurasian Journal of Analytical Chemistry 5(2),pp.187-203,2010.
[4] X. Yuanzhe, Z. Hongjing, Li Bo, Zhao Xin," Study on Detection of SF_6 Gas Leakage Based on Photoacoustic Spectrometry", International Conference on Measuring Technology and Mechatronics Automation, pp.261-265,2009.
[5] J. BESSON,"Photoacoustic spectroscopy for multi-gas Sensing using near infrared lasers", Ph.D thesis, À La Faculté sciences ET Techniques de L'ingénieur, Lausanne, 2006.
[6] M. Mattiello," Gas Traces Measurement by Photoacoustic Spectroscopy Using Helmholtz Resonator-Based Sensors",Ph.D thesis, À La Faculté Des Sciences ET Techniques de L'ingénieur, Lausanne, 2008.
[7]N. Mingsheng,L. Qiang,L. Kun,Y. Yiqian and G. Xiaoming," Temperature-dependent photoacoustic spectroscopy with a T shaped Photoacoustic cell at low temperature", Optics Communications287, pp.180–186, 2013.
[8]N. Mingsheng,L. Qiang,L. Kun,Y. Yiqian and G. Xiaoming, " Temperature-dependent characteristics of a photoacoustic detector", Optik, No. of Pages 5,2012.
[9] G.R. Lima, M.S. Sthel, M.G. da Silva, D.U.S. Schramm, M.P.P. de Castro and H. Vargas," Photoacoustic spectroscopy of CO_2 laser in the detection of gaseous molecules", Journal of Physics: Conference Series274,2011.
[10]Y. Yun, W. Chen, Y. Wang and C. Pan, "Photoacoustic detection of dissolved gases in transformer oil", European Transactions on Electrical Power, 18, pp.562–576, 2008.
[11] R. Cernat, C. Matei, A.M. Bratu, C. Popa, D.C.A. Dutu, M. Patachia, M. Petrus, S. Banita and D.C. Dumitras," Laser Photoacoustic Spectroscopy method for measurements of trace gas concentration from human breath", Romanian Reports in Physics, Vol. 62, No. 3, pp .610–616, 2010.
[12] D.C. Dumitras, D.C. Dutu, C. Matei, A. - M. Magureanu, M. Petrus and C. Popa," Improvement of A Laser Photoacoustic Instrument for Trace Gas Detection", ISSN, Series A, Vol. 69, No. 3, pp. 45-56, 2007.
[13] J. Lia, X. Gaoa, L. Fanga, W. Zhanga and H. Cha," Resonant photoacoustic detection of trace gas with DFB diode laser", Optics & Laser Technology 39, pp. 1144–1149,2007.
[14] A. Tavakoli, M. Tavakoli, M. Taheri, and H. Saghafifar," Design, Simulation and Structural Optimization of a Longitudinal Acoustic Resonator for trace gas detection using Laser Photoacoustic Spectroscopy (LPAS)", International Conference on Optics and Photonics, CSIO, Chandigarh, India, 30 Oct.-1 Nov. 2009.

التقنية الضوئية الصوتية للكشف عن الغازات

أسراءعامركاظم **محمدعبدالاميرحسين**
قسم هندسه الليزروالالكترونيات البصرية/ جامعة النهرين

الخلاصه:
مطيافية الليزر الضوئيه-الصوتيه هي احدى التقنيات لكشف وقياس اثار الغازات بأستخدام اشعه الليزر تحت الحمراء. تستند هذه التقنيه على توليد موجه صوتيه في غاز مستثار بوساطه شعاع ليزر مضمن لطول موجي يتطابق مع خط الامتصاص لنوع الغاز وبتردد للتضمين يتوافق مع المرنان الصوتي المستعمل، و يستخدام مايكروفون حساس للكشف عن هذه الموجة الصوتية.
يمثل هذاالعمل طريقه عمليه للكشف عن الغازات باستخدام الليزر في التقنية الضوئية-الصوتية حيث أستخدم غاز الفريون-12 كنموذج.
تم انجاز التجارب العملية بتصنيع خلية ضوئية-صوتية تتألف من مرنان صوتي بطول 50ملم وبقطر4 ملم تمثل نهايتاه محجرين للغاز باستعمال نوافذ ذات سماحيه جزئيه لاشعاع µm10.6 لإحكام فتحات النهايتين. وقد أستخدم لاقط صوتي (مايكروفون) حساس تم تثبيته في منتصف المسافة بين المرآتين للكشف عن الاشاره الصوتيه.
استعمل في هذا البحث ليزر ثنائى أوكسيد الكاربون وبقدرة مؤثرة بين 0.7 و 2واط لتوليد الموجه الصوتيه.
وقد تم انجاز تحليل البيانات العملية باستخدام برامجية (Matlab).