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Pulsed photoacoustic gas cell design for low pressure studies

M.D. Rabasović [b,*], J.D. Nikolić [a], D.D. Markushev [b], J. Jovanović-Kurepa [b]

[a] *Institute of Nuclear Sciences "Vinca", P.O. Box 522, 11001, Serbia*
[b] *Institute of Physics, Pregrevica 118, 11080 Beograd – Zemun, Serbia*

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Abstract

The designing of the photoacoustic cell is critical to obtain rather good signal to noise ratio. All theoretical and experimental studies predict that for a constant light absorption, the amplitude of the photoacoustic signal reaching the microphone depends on the size of the cell and in general increases as cell dimensions are reduced. For constant laser energy and beam – cell geometry, amplitude of photoacoustic signal also depends on sample pressure. Such an effect is obvious in the low pressure region, where collisionally induced processes are dominant. Functional amplitude vs. pressure behavior is not a simple one, and must be established for any type of gas samples. Here we would like to emphasize that our attention was dedicated strictly to low gas mixture pressure region (less than 100 mbar), where some collisionally induced relaxation processes can be monitored by analyzing photoacoustic signal shape and amplitude behavior as a pressure dependent quantities.

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1. Introduction

In recent time the atmospheric pollution has become an issue of a great concern. It has been recognized that even trace concentrations of some atmospheric molecular and atomic species can have a substantial impact in diverse areas [1,2]. In the past, numerous techniques have been developed and successfully applied to the trace gas monitoring. Some of them have become standard methods and are widely used on a routine basis [3]. One of them is pulsed infrared photoacoustics spectroscopy (PAS), which is used not only for precise detection and measurements of minimal trace gas concentrations in the atmosphere (minimum and maximum concentrations extend into the parts-per-trillion to percent ranges) but for their intensive investigation on atmospheric and subatmospheric pressures in order to reach the thorough understanding of complex physical and chemical processes and interactions involved [4–6]. Here, we used SF_6 as an investigated absorber, mixed with different buffer-gases (Ar and N_2), in order to analyze and understand its physical characteristics through photoacoustic signal shape investigation and behavior under the different gas mixture pressures and strong laser radiation field–molecule interactions.

The photoacoustic spectroscopy is based on the sensitive detection of acoustic waves launched by the absorption of pulsed or modulated laser radiation via the transient localized heating and expansion in an investigated gas sample. This effect is caused by the transformation of at least part of the excitation energy into kinetic (translational) energy by energy exchange processes. The designing of the photoacoustic cell is really critical to obtain rather good signal to noise ratio [3,7–9]. Photoacoustic cell serves as a container for the gas sample and the microphone (usually used for generated acoustic wave detection). An optimum cell design represents a crucial point for different applications. Many cell configurations have been presented including acoustically resonant and nonresonant cells, single- and

* Corresponding author. Tel.: +381 11 31 60 260; fax: +381 11 31 62 190.
E-mail address: rabasovic@phy.bg.ac.yu (M.D. Rabasović).

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multipass cells, as well as cells placed intracavity [2,3]. Our cell, presented here, is a large cell adopted for pulsed excitation and low pressure investigation [9].

2. Experimental set-up and photoacoustic cell design

The experimental set-up is given in Fig. 1 [9]. TEA CO_2 laser was used as a light source of 45 ns FWHM pulse with 2 μs tail and the top hat profile. The spatial profile was defined by the iris. The laser beam was not focused. This is the typical set-up for photoacoustic gas samples studies, and numerous different versions and modifications of the general scheme have been discussed in the literature [2,3].

The stainless-steel photoacoustic cell was utilized, which had an overall length of 18.5 cm and diameter of 9.3 cm, having specific T-cell form (Fig. 2). A capacitive microphone (Knowles Electronic Co., model 2832, 4 mm diameter) was mounted in the cell at the distance of 4.7 cm from the entrance window. Two NaCl windows were attached in the two ports of the three-port photoacoustic cell, while the third port had quartz optical window. A high vacuum pump system was utilized for the cell evacuation, achieving the background pressure of 10^{-6} mbar. A ceramic capacitance manometer (CCM Instruments, model 501) with the accuracy of 0.1% was installed to monitor gas pressure. Direct calorimetric determination of the laser energy and energy absorbed in the cell was performed by a calibrated Gen-Tec ED-20C pyroelectric joulemeter.

Both, the acoustic waveform and the laser excitation energy were digitized by a waveform recorder (Keithley MetraByte PCPI Scope plug-in board). Data acquisition and processing were performed by a personal computer. Acoustic waveforms were stored for subsequent analysis. The analysis includes the calculation of the photoacoustic signal (based on the intensity of the first peak) normalized to laser excitation energy.

We examined SF_6–Ar gas mixtures. Measurements were performed at the mixture pressure range of (10–100) mbar and fluence range of (0.4–1.5) J/cm^2. Absorber pressure (SF_6) was kept constant on 0.47 mbar. Mixture pressure was changed by changing the buffer gas (Ar) pressure. Main parameter measured directly in our experiment is photoacoustic (PA) signal. This signal corresponds to the

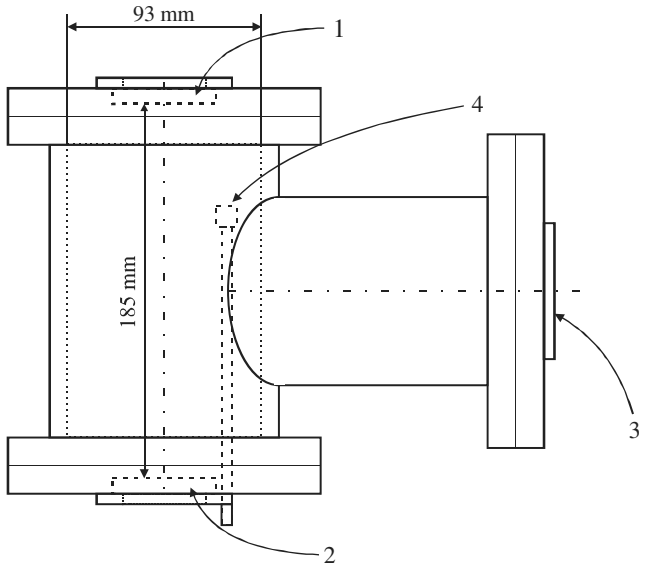

Fig. 2. Photoacoustic cell used in our experiment, 1 – entrance NaCl window, 2 – exit NaCl window, 3 – quartz window, 4 – microphone.

acoustic wave (pressure change) detected by microphone. We said earlier that photoacoustic cells for pulsed light sources can be large or small, depending on their capability to obtain individual PA signals. In this experiment we can obtain and see individual PA signals, and our cell can be considered as a large one. The fact that cell is large enough gives us a possibility to detect clear photoacoustic signals without interferences from the cell walls and windows. In this case, the cell can be considered as infinitely large and boundary conditions become irrelevant. This enables straightforward calculation of photoacoustic signals [9].

The type of measurements presented here, based on low pressure studies of collisionally induced processes during the nonlinear (multiphoton) molecular absorption and relaxation, sets a challenging standard to the detection and monitoring of trace gases. Ideally a detection technique (photoacoustics in our case), should fulfill the following requirements: (a) feasibility of detecting numerous compounds with one instrument; (b) high sensitivity in order to permit the detection of very low concentrations; (c) high selectivity in order to differentiate between different

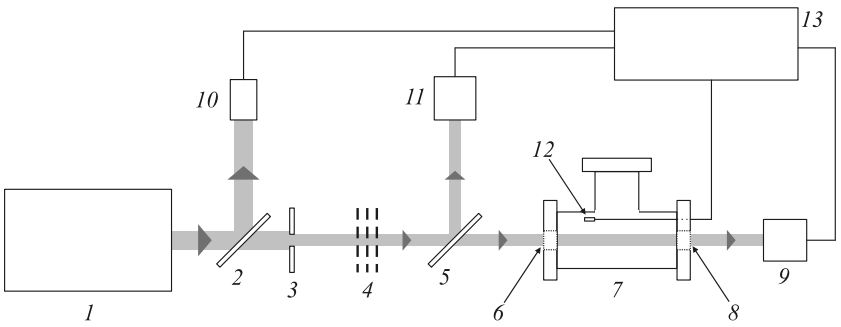

Fig. 1. The experimental set-up: 1 – TEA CO_2 laser; 2, 5 – beam splitters; 3 – iris; 4 – attenuators; 6, 8 – windows; 7 – photoacoustic cell; 9, 11 – joulemeters; 10 – photon drag detector; 12 – microphone; 13 – data acquisition system.