

Photoacoustics: an acoustic method for trace gas measurements

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Introduction

The photoacoustic (PA) effect, the generation of sound by the absorption of pulsed or modulated light, has been known since the famous experiments of Alexander Graham Bell in 1880. Scientific and practical applications of the photoacoustic technique have been considerably promoted by the invention of laser, which is an ideal light source for photoacoustics. Recently, photoacoustic detectors play an increasing role in trace gas detection and analysis. The sensitivity of a small, simple and cheap photoacoustic detector is only slightly lower than that of a very sophisticated and expensive optical system equipped with a multipass optical absorption cell and a liquid nitrogen cooled infrared detector.

The performance of a photoacoustic trace gas detector depends mostly on its acoustical design. However, the majority of the published or commercially available photoacoustic detectors are acoustically not optimized. Considerable improvement in the sensitivity could be achieved by using better acoustic designs and more sophisticated measurement and data evaluation methods. The PA research at the Fraunhofer Institute of Building Physics is targeted to the development of specific, acoustically optimized PA detectors for each important application field of the PA technique, such as trace gas measurement and analysis in air; emission control of exhaust gases of vehicles and industrial combustion; detection of poisonous gases, drugs and explosives, monitoring and control of process gases, etc.

Photoacoustic signal generation

When a medium is illuminated by a laser beam, the light energy absorbed in the medium will be transformed to heat. The details of the (in some cases quite complicated) absorption and relaxation processes are not the subject of the present discussion. The relevant effect is a local heating of the medium. As a result the temperature will increase and mechanical stress (pressure) will be generated in the illuminated volume. In the case of stationary illumination an equilibrium temperature distribution will be achieved, while the mechanical stress will be relaxed by thermal expansion. In a gas confined in a closed container the stationary absorption of light will result in an increase of the average temperature and pressure.

The situation will be completely different, if the illumination is time dependent, i.e., pulsed or modulated laser is used. The absorption of this kind of beam generates simultaneously a thermal and a sound wave. However, the propagation distance of the thermal wave is very small. For 1 kHz modulation frequency the thermal wavelength is smaller than 0.5 mm in air, and the amplitude decays to

1/500 of its original value within this distance. Therefore, the contribution of the thermal wave can be omitted, if the distance of the microphone from the laser beam exceeds the thermal wavelength.

Sound wave generation can be described by the acoustic wave equation with a thermal source term. This equation can be written as follows [1, 2]:

$$\partial_t^2 p - c^2 \Delta p = (\gamma - 1) \partial_t h \quad (1)$$

where p , c , γ , and h are the pressure, sound velocity, adiabatic constant of the gas, and the heat power density, respectively. The nature of the source term is not specified. In fact, any kind of physical process that can deposit heat energy locally and transiently into the medium can produce sound.

The source term on the right hand side of eq.1 can be written as

$$-(\gamma - 1) \partial_t \operatorname{div} \mathbf{I} \quad (2)$$

where \mathbf{I} is the intensity vector of the light beam. Since the source term is proportional to the time derivative of the divergence of the light intensity vector \mathbf{I} , sound can be generated only by nonstationary and inhomogeneous illumination.

Sound generation in a PA detector

Assume that a laser beam passes through a cylindrical cell with much smaller dimensions than the acoustic wavelength and that the cell is closed by entrance and exit windows with negligible absorption. The PA cell is filled with a weakly absorbing gas. The sound pressure amplitude P generated by the PA effect in this simple PA cell can be given as

$$P = \frac{(\gamma - 1) W}{\omega V} \alpha l \quad (3)$$

where W , α , l , ω and V are the light power, absorption coefficient, length of the cell, (angular) modulation frequency and cell volume, respectively [1].

Signal enhancement can be achieved by a resonant PA detector. In this case the detector contains an acoustic resonator, and the modulation frequency of the laser is tuned to a selected resonance of the resonator. Best performance can be achieved by proper design of the light path geometry and the position of the microphone.

The dynamic range of the PA detector is determined mostly by the microphone. The linear dynamic range of a PA detector for the absorption coefficient (or concentration)

usually exceeds 4 – 6 orders of magnitude; therefore the PA detector can be calibrated by a two point calibration measurement.

Sensitive PA detector designs

The most sensitive PA device to date, a differential PA detector has been developed [3] in Heidelberg (Fig 1).

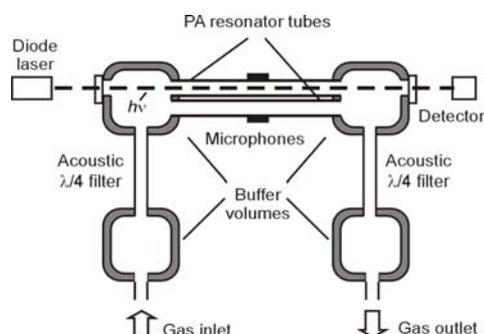


Figure 1: Schematic drawing of the differential PA detector with two resonator tubes, buffer volumes and acoustic $\lambda/4$ filters. The first longitudinal acoustic mode with a Q factor of ~ 31 is excited.

Two identical acoustic resonators (open tubes) are placed in the middle of the detector chamber. Windows are separated by quarter-wavelength buffers, gas in- and outlets are coupled to the buffers through a quarter-wavelength tube and a second buffer. This acoustic filter system suppresses acoustic and flow noise very effectively. The resonator tubes are electrically isolated from the metal housing. The two microphones are placed in the middle of the resonator tubes. The same noise and background signal is recorded in both resonators, while the PA signal is only generated in one of them. The difference of both microphone signals contains the whole PA signal without noise.

The sensitivity can be further increased by using an optical multipass arrangement within the PA detector [4]. However, this detector can be used only with wavelength modulation of the laser, because by intensity modulation a background PA signal is generated at the surfaces of the built-in spherical mirrors. Nevertheless, a sensitivity increase of a factor of ten was achieved.

A similar idea was applied for the development of an acoustically open PA detector [5]. In this case the PA signal is generated in the open air by a multipass optical arrangement, and the acoustic signal is concentrated on the membrane of a miniature microphone. This patented system is designed for monitoring trace concentrations of unstable molecules or strongly polar molecules in the air.

Possible optimization of the PA sensors

For the best performance of a PA sensor it is not enough to optimize its different subsystems separately; the interaction of every single part of the system has to be considered. This means that

- acoustical system (resonator(s) and filters),
- optical system (multipass arrangement built in the acoustic cell),

- the method of signal generation (pulsed or modulated excitation, modulation method, frequency, pulse duration, etc.),
- the gas flow system and the signal acquisition and evaluation system

should be optimised matched to each other simultaneously for best performance. Different applications may require different PA sensor designs, thus the optimization and matching procedure has to be performed for each important application fields.

The main directions of the planned development and optimization are:

- reduction of the size of the differential PA detector,
- development of fiber coupled single and multipass PA sensors,
- application of high-Q acoustic resonators with pulsed excitation,
- development of open PA sensors for high temperature (up to 400 °C) applications,
- development of PA sensors with broadband IR-emitters.

Conclusion

The photoacoustic technique is a powerful tool for sensitive and selective trace gas measurement and real-time analysis of gas components. Thereby the acoustical subsystem is one of the main parts of the arrangement. Because of this, the main effort in the development of better photoacoustic sensors should be concentrated on the acoustic design. This provides high sensitivity PA sensor products for diverse applications.

References

- [1] A. Miklós *et al.*: Experimental and Theoretical Investigation of Photoacoustic Signal Generation by Wavelength-Modulated Diode Lasers, *Appl. Phys. B* **58** (1994), 483-492
- [2] S. Schaefer, A. Miklós, and P. Hess: Quantitative signal analysis in pulsed resonant photoacoustics, *Appl. Opt.* **36** (1997), 3202-3211
- [3] A. Miklós and P. Hess: "Modulated and pulsed photoacoustics in trace gas analysis", *Anal. Chem.* **72** (2000) 30A-37A
- [4] J. Ng, A.H. Kung, A. Miklós and P. Hess: Sensitive wavelength-modulated photoacoustic spectroscopy with a pulsed optical parametric oscillator, *Opt. Letters* **29**, (2004), 1206-1208
- [5] A. Miklós, S.-C. Pei, and A.H. Kung: Multipass acoustically open photoacoustic detector for trace gas measurements, *Appl. Opt.* **45** (2006), 2529-2534