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Multi-resonator photoacoustic spectroscopy

Kun Liu [a,*], Jiaoxu Mei [a], Weijun Zhang [a], Weidong Chen [b], Xiaoming Gao [a,*]

[a] Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Hefei 230031, China
[b] Laboratoire de Physicochimie de l'Atmosphère, Université du Littoral Côte d'Opale, 189A, Av. Maurice Schumann, 59140 Dunkerque, France

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ABSTRACT

In this letter, we report on the development of an innovative multi-gas sensor based on multi-resonator photoacoustic spectroscopy (MR-PAS). This novel technique offers multi-laser operation to simultaneously monitor multiple pollutant species using a single photoacoustic spectrophone. A photoacoustic cell including three acoustic resonators operating at different resonant modes was designed, a single microphone was used to listen the photoacoustic signal in each resonator simultaneously. Feasibility and performance of the innovated MR-PAS sensor was demonstrated by simultaneous trace gas detection of H_2O vapor, CH_4 and CO_2 using three near infrared distributed feedback diode lasers. 1σ normalized noise equivalent absorption coefficients (NNEA) of 2.1×10^{-9} cm^{-1} W/Hz$^{1/2}$, 2.9×10^{-9} cm^{-1} W/Hz$^{1/2}$ and 6.5×10^{-9} cm^{-1} W/Hz$^{1/2}$ were respectively achieved for H_2O, CH_4 and CO_2 detection at normal atmospheric pressure.

1. Introduction

Photoacoustic spectroscopy (PAS) is a sensitive, selective and well established method for trace gas detection which has been successfully employed in numerous applications. Amid spectroscopy-based optical sensors, PAS based on photo-acoustic convention effect offers several intrinsic attractive features including ultra-compact size, free from cross-response of light scattering, zero-background and broad band wavelength-independent acoustic signal measurements (free of wavelength-dependent photo-detector). Up to date, various concepts have been explored with respect to PAS-based sensing platform, such as resonant PAS, cantilever enhanced PAS and quartz enhanced PAS [1–7].

There is strong need for the development of PAS sensor capable of multi-wavelength and multi-component detection. One such application, the original motivation for the present work, is the simultaneous measurement of several green house gases and aerosol optical absorption. However, most of the reported PAS sensors are not able to offer multi-component detection capability, except for using widely tunable laser sources. In fact, narrow tuning range of most commercially available lasers make multi-component detection needing multi-laser sources. For example, recently, H. Wu et al. reported a dual-gas QEPAS sensor by exciting two different quartz tuning fork resonance modes with two lasers [8]. In conventional PAS configuration, a single acoustic resonator operates at its own resonant frequency f_0 and it is difficult to simultaneously retrieve the PAS signals resulting from different lasers for multi-component analysis (in this case, monitoring of multi-gas should be made by consequently switching corresponding laser source).

2. Experimental

As an alternative method, we developed an innovative multi-resonator phtoacoustic spectroscopy (MR-PAS) scheme for simultaneous monitoring of multi-component with multi-laser coupled to multi-resonator in a single phtoacoustic spectrophone. Geometric configuration of the MR-PAS cell is shown in Fig. 1. The MR-PAS cell includes three cylindrical acoustic resonators with their own specific lengths offering different acoustic resonant frequencies. These acoustic resonators were set in a cylinder with a diameter of 50 mm and distributed in a circle of 25 mm diameter, as shown in Fig. 1. The inner diameter of each resonator was 10 mm. The length of acoustic resonator 1 (indicated as AR1), acoustic resonator 2 (AR2) and acoustic resonator 3 (AR3) was 120 mm, 110 mm and 100 mm, respectively, such that the resonant frequency of each resonator was separated by a gap of ~100 Hz that was larger than a FWHM (full width at half maximum) of the resonance profile of a typical cylindrical resonator with a length of 100 mm, which ensures no signal cross-talking during phase-sensitive signal demodulation for each resonator. A buffer volume was set at both sides of the cylinder, and the total length of the PAS

* Corresponding authors.
E-mail addresses: liukun@aiofm.ac.cn (K. Liu), xmgao@aiofm.ac.cn (X. Gao).

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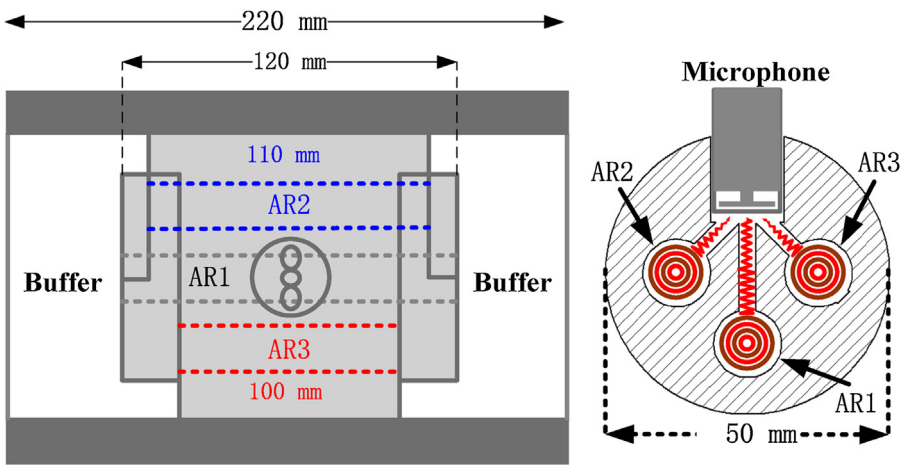

Fig. 1. Sectional view of the developed MR-PAS sensor: geometric configuration of multi-resonators PAS cell (left) and geometric distribution of three multi-resonators integrated in a cylinder (right).

Table 1
Summarized parameters of the developed MR-PAS sensor.

Parameters	Acoustic resonator		
	AR1	AR2	AR3
Length, mm	120	110	100
Resonant frequency, Hz	1400	1510	1610
Molecule	CO_2	CH_4	H_2O
Target line, cm^{-1}	4989.97	6046.95	7161.41
Laser wavelength, μm	2.004	1.653	1.396
Laser power, mW	1.5	8.6	7.8
NNEA, $cm^{-1}W/Hz^{1/2}$	6.5×10^{-9}	2.9×10^{-9}	2.1×10^{-9}

cell was 220 mm. A hole with an inner diameter of 3 mm was set in the middle of each resonator and used as a sound wave guide tube between acoustic resonator and microphone detector. Such a design allows for the detection of PAS signal in each resonator using a single microphone, as illustrated in Fig. 1. The signal from each resonator was demodulated at its own resonant frequency using a lock-in amplifier.

Validation of the developed MR-PAS sensor has been carried out by the measurements of three important green house gases: H_2O vapor, CH_4 and CO_2. Selection of the absorption lines for each species was made based on the HITRAN04 database [9]. The selected interference-free lines are given in Table 1. The line for CH_4 detection is a R_3 triplet consisting of 3 lines: F_1, F_2 and A_2, overlapped and unresolvable even at low pressure under Doppler broadening conditions.

A schematic of the MR-PAS experimental setup is shown in Fig. 2. Three fiber-coupled distributed feedback (DFB) diode lasers operating at 1.396 μm (NEL), 1.653 μm (NEL) and 2.004 μm (eblana photonics) have been used for detection of H_2O vapor, CH_4 and CO_2, respectively. Three commercial diode laser controllers (ILX Lightwave LDC-3724) were used for laser current and temperature controls. Laser wavelength scans were realized by feeding an external voltage ramp from a function generator (SPF05, NANJING SAMPLE INSTRUMENT) to the laser diode current which swept the laser wavelengths back and forth across the absorption feature of each target molecule at a rate of 1 Hz, simultaneously. Wavelength modulation and second harmonic detection was employed in the present work for sensitive trace gas detection. The wavelength modulation was achieved by adding a sine wave to the DFB laser diode current. The sine form wave was supplied by the sinusoidal signal output of a lock-in amplifier (Stanford Research Systems, Model SR 830 DSP). The voltage ramp and the sine wave were combined with a home-made adder and then fed to the laser controller. Acoustic signals from each resonator were detected with the same microphone (BSWA, MP201) and demodulated by using three lock-in amplifiers separately. A unique time constant of the lock-in amplifier of 10 ms in combination with an 18 dB/octave slope (leading to a detection bandwidth of 9.375 Hz) was used. The demodulated signals were subsequently digitalized with a DAQ card (NI-USB-6212) and displayed on a laptop via a LabVIEW interface. The used acoustic resonators and their corresponding specific geometric parameters are given in Table 1.

3. Results and discussion

Characteristics of each acoustic resonator were experimentally investigated to determine the specific resonant frequency for each resonator at atmospheric environment. Measurements of H_2O vapor absorption signal using a 1.396 μm laser was performed for this study. Fig. 3 shows the square of the signal amplitude as a function of frequency. The data were fitted to a Lorentz contour, which describes power in a classical driven oscillator as a function of frequency. From the results shown in Fig. 3, optimum resonant frequencies f_{10} = 1400 Hz, f_{20} = 1510 Hz and f_{30} = 1610 Hz were found for resonators AR1, AR2 and AR3, respectively. These values correspond to the first longitudinal resonant mode of each resonator [1].

To evaluate the capacity of multi-gas detection of the developed MR-PAS proof-of-concept, a mixture of 610 ppm CH_4, 1430 ppm H_2O vapor and 7042 ppm CO_2 in N_2 and air was filled in the MR-PAS sensor at atmospheric pressure. The concentration of each molecule in the mixture was determined with direct absorption spectroscopy. The lasers emitting at 2.004 μm (CO_2), 1.653 μm (CH_4) and 1.396 μm (H_2O) were modulated at 700 Hz (half of the optimum frequency, $f_{10}/2$), 755 Hz ($f_{20}/2$) and 805 Hz ($f_{30}/2$), respectively. Modulation amplitude applied to each laser was optimized by investigating detected signal amplitudes at different modulation amplitudes. Fig. 4(a) depicts the 2f-signals of H_2O, CH_4 and CO_2 obtained simultaneously using optimum modulation amplitudes (850 mV, 600 mV and 800 mV for H_2O, CH_4 and CO_2, respectively, The 2f-signal of H_2O was divided by 10). The noise level, defined as standard deviation in the baseline fluctuation of a non-absorption portion in the 2f-signal spectrum, was found to be ~20 μV for all resonators. The minimum detection limit were then estimated to be 1.3 ppm for H_2O, 4.4 ppm for CH_4 and 140 ppm for CO_2, respectively. Taking into account the used lasers power and the detection bandwidth of 9.37 Hz (10 ms time constant combined with an 18 dB/octave slope filter), 1σ normalized noise equiva-