

# A continuous-wave optical parametric oscillator for mid infrared photoacoustic trace gas detection

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**Abstract:** We present a continuous-wave, pump-resonant, singly-resonant optical parametric oscillator based on periodically poled lithium niobate in a linear dual-cavity design which is applied for photoacoustic trace gas detection between 2.35 and 3.75  $\mu\text{m}$ . Pumped by a Nd:YAG laser at 1064 nm a single-frequency output power of 2 x 100 mW is achieved. The frequency tuning qualities of the OPO allow to scan gas absorption structures. An ethane detection limit of 110 ppt is achieved. Sensitivity, tuning qualities and compact design make this OPO an ideal laser source in a transportable photoacoustic spectrometer.

**OCIS codes:** (190.4970) Parametric oscillators and amplifiers; (300.6340) Spectroscopy, infrared; (120.6200) Spectrometers and spectroscopic instrumentation

## 1. Introduction

Laser spectroscopy is a powerful tool for monitoring trace amounts of volatile organic compounds in biology, atmospheric chemistry and medicine. These molecules possess strong absorption fingerprint spectra in the 3 $\mu\text{m}$  wavelength region, where photoacoustic [1] and cavity-leak-out spectroscopy (CALOS) [2], a continuous-wave variant of cavity ring-down spectroscopy (CRDS), have been established as methods with high sensitivity, selectivity and time resolution. Besides difference-frequency generation (DFG) [3] as light source, singly-resonant optical parametric oscillators (SRO) using periodically poled lithium niobate (PPLN) as nonlinear medium offer very good frequency tuning potential and higher conversion efficiencies. High pump powers are needed, however, to reach threshold. Herpen *et al.* used a 15 W cw 1064 nm pump laser to achieve 2.2 W cw 3 $\mu\text{m}$  idler power [4]. Threshold can be lowered by pump resonance. With a common-cavity SRO design (pump and signal resonated in one cavity) without etalon [5] threshold was reduced to 270 mW. This OPO was applied for trace gas detection using CALOS. Tunability was limited by mode hops making it impossible to tune into maximum of absorption. Here we present a dual cavity design (pump and signal resonated in separate cavities) with intracavity etalon, solving these frequency tuning problems. This new OPO is applied in a transportable photoacoustic spectrometer.

## 2. OPO

The continuous-wave, pump-resonant, singly-resonant optical parametric oscillator (cw-PR-SRO) [6] is set up in a linear dual-cavity design (Fig.1.) and uses periodically poled lithium niobate (PPLN) as nonlinear medium. The crystal contains 33 gratings with poling periods between 28.4 and 30.96  $\mu\text{m}$  to cover the wavelength range between 2.35 and 3.75  $\mu\text{m}$ . The first crystal surface is HR coated for both

pump and signal waves. The pump-cavity is build by a meniscus mirror (antireflection coated for signal and idler) followed by the concave signal-cavity mirror. Both mirrors are attached to piezoelectric transducers to adjust cavity lengths. An intracavity etalon (YAG, 0.5mm, 50 % signal reflectivity coating) suppresses spontaneous mode hops.

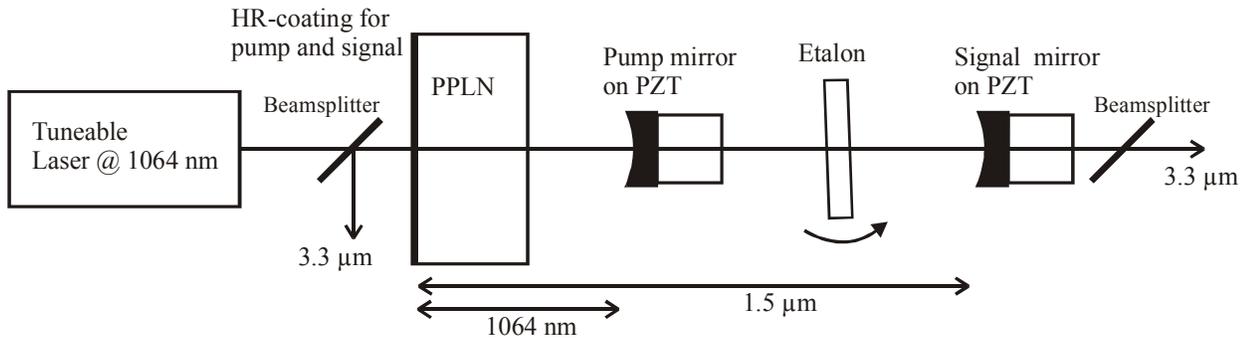


Fig. 1. Linear cw-PR-SRO setup

The pump enhancement lowers the external pump threshold down to 380 mW (Fig. 2.). Using a 2.5 Watt Nd:YAG pump laser (1064 nm, Innolight) a maximum idler output power of  $2 \times 100$  mW is achieved. The dual-cavity design enables the combination of low threshold with high tunability. The pump-cavity is locked to the laser (Pound-Drever-Hall), the signal-cavity is locked to maximum idler power. Mode-hop-free operation was observed over 20 minutes. During this time the idler frequency stability was better than the  $\pm 30$  MHz accuracy of our wavemeter (WA 1500, Burleigh).

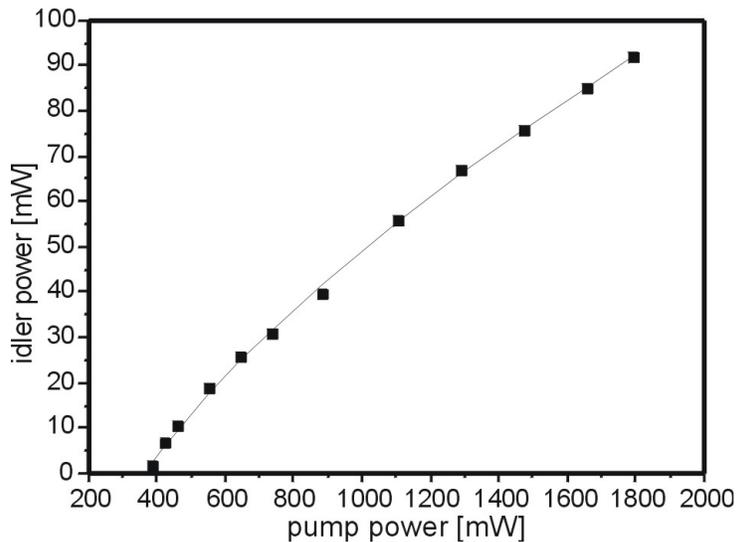


Fig. 2. Single end idler output power versus incident pump power

Idler frequency tuning is performed in four steps:

1. Coarse tuning between 2.35-3.75  $\mu\text{m}$  by selecting one of the 33 crystal-gratings via motor driven translation stage.
2. Temperature tuning of the PPLN crystal between 150-190°C
3. 450 MHz steps mode hop tuning (54 GHz in total) by turning the etalon via galvanometer.
4. Fine tuning (1.5 GHz mode hop free, 30 GHz in total) by tuning the pump laser (Fig. 3.).

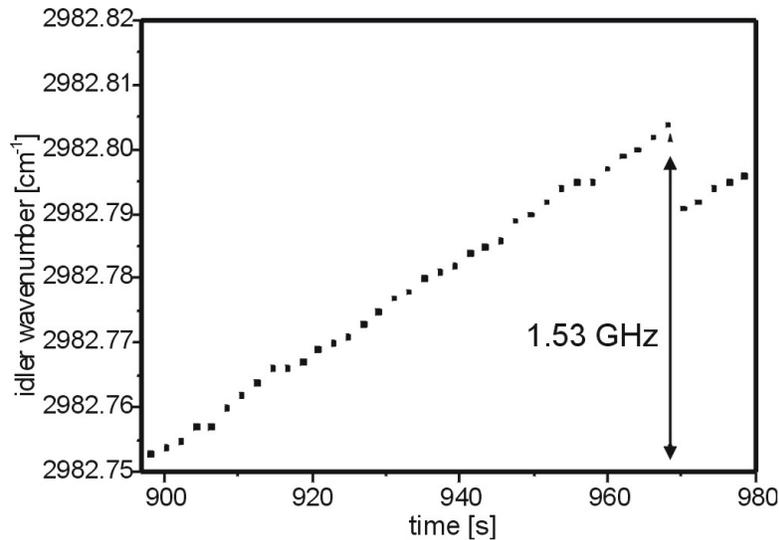


Fig. 3. Idler fine tuning by tuning of the pump laser

### 3. Photoacoustic spectroscopy

The photoacoustic spectrometer consists of the OPO, chopper, photoacoustic cell and pyroelectric detector and is installed on a 120 x 75 cm bread board. The amount of radiation absorbed by the molecules is measured by its conversion into heat. The 3  $\mu\text{m}$  laser beam is modulated in amplitude at the resonance frequency of the photoacoustic cell, yielding a standing acoustic wave. The signals from the microphone and the pyroelectric detector behind the cell are processed with two lock-in amplifiers (Stanford Research, SR 830). The tunability of the OPO allows to scan the strongest gas absorption structures in the 3  $\mu\text{m}$  region. At atmospheric pressure ethane and ethylene show absorption structures between 0.1 and 1  $\text{cm}^{-1}$  broad. These features can be covered using the 450 MHz steps etalon tuning. In fig. 4 a scan of the  $^{\text{P}}\text{Q}_1$  sub branch of ethane is compared to an accurately determined absorption spectrum (FTIR [7], CALOS [5]) showing good agreement (Fig.4.). With pure carrier gas in the photoacoustic cell background signals due to window absorptions are present. At 10 seconds lock in time, the noise level of this background corresponds to a minimum absorption coefficient of  $3.2 \times 10^{-9} \text{ cm}^{-1}$ , yielding a detection limit of 110 ppt for ethane. For the 635 ppb ethylene peak we found deviations from the HITRAN [8] calculation (Fig.5): In the measured spectrum the absorption is about 25% smaller than the simulated data, and the absorption features are more pronounced. The latter indicates, that the ethylene pressure broadening coefficients used in the HITRAN database are too large.

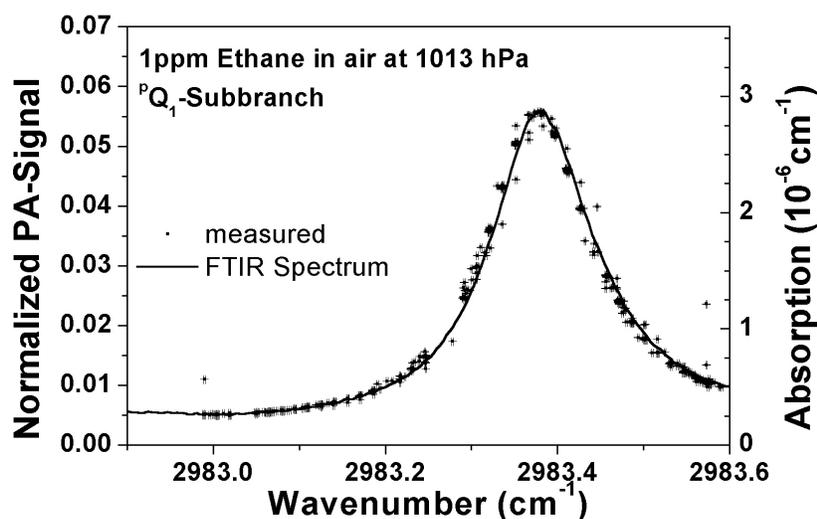


Fig. 4. Scan over an ethane absorption peak (atmospheric pressure, ethane concentration 1ppm) with 450 MHz etalon mode hop tuning

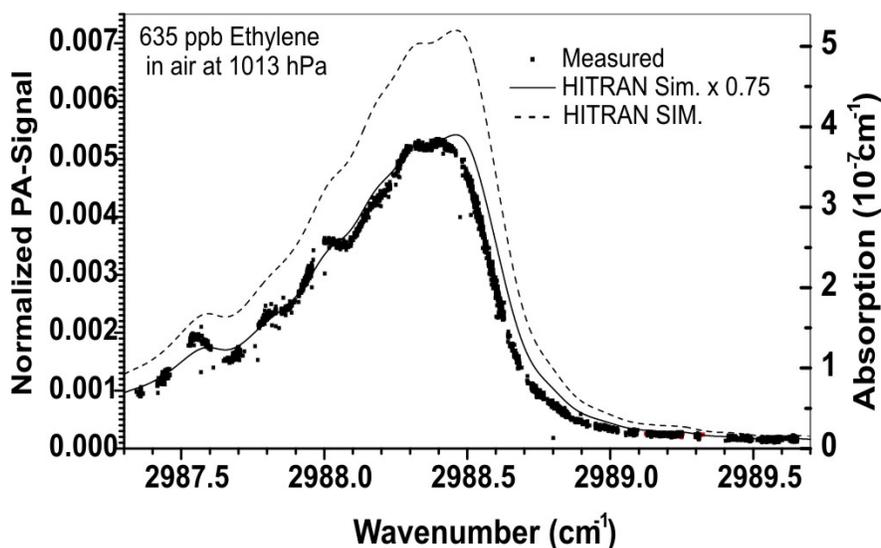


Fig. 5.  $2\text{ cm}^{-1}$  wide scan over an ethylene absorption peak (atmospheric pressure, ethylene concentration 635 ppb) with 450 MHz etalon mode hop tuning

#### 4. Summary

The dual-cavity PR-SRO combines low oscillation threshold with wide tunability. It allows to scan molecular absorption structures at atmospheric pressure. With 70 mW available for photoacoustic spectroscopy we reach an ethane detection limit of 110 ppt. The results are an important step towards a fully automated, transportable spectrometer for long term monitoring as it is in routine operation for ethylene detection using  $\text{CO}_2$  lasers [9].

## 5. Acknowledgements

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