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Narrow linewidth optical parametric oscillator for photoacoustic spectroscopy.

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Nanosecond optical parametric oscillators (OPO) show unique features of wavelength tunability. They can be used in particular for the spectroscopy of many molecules in the range of 3.3 μm - 3.4 μm (C-H bond absorption lines). They are well suited for LIDAR and sensing applications such as photoacoustic spectroscopy.

To reduce the natural linewidth of our LiNbO₃ OPO (1 cm^{-1}), we have inserted a Fabry-Pérot interferometer (FPF) into the cavity. The thickness is optimised so that only one of its resonances lays into the OPO natural linewidth $\Delta\nu$. The width of the resonance $\Delta\nu_r$ is related to the mirrors reflectivity. It is chosen to match the emission linewidth requirement. The spacing between the FPF mirrors is tunable around 1 mm thanks to a piezoelectric stack. This allows a fine tuning of the OPO emission frequency. The mirror reflectivity is 70 %. It gives a good trade-off between the FPF spectral filtering and its transmission at resonance. To measure the signal spectral linewidth, interference rings are produced with a 0.4 cm^{-1} free spectral range Fabry-Pérot spectrometer. They are monitored on a Vidicon tube camera (fig 1) and show that we are able to decrease the signal spectral linewidth down to 0.08 cm^{-1} .

The absorption spectrum of CH₄ is monitored by tuning the idler wavelength of the OPO around 3.380 μm . The theoretical absorption spectrum at atmospheric pressure has a width of 0.3 cm^{-1} and shows a double peak structure. The infrared beam is sent into a photoacoustic cell which is filled with a mixture of 18 ppm of CH₄ in artificial air. The light absorbed by the gas generates an acoustic signal which is registered by a microphone. The absorption line can be clearly monitored (fig 2). Two resonances can be observed corresponding to two successive orders of the FPF separated by 5 cm^{-1} . The double peak structure of the line is resolved. This measurement shows that the idler beam linewidth is significantly smaller than 0.3 cm^{-1} .

The sensitivity of the measurement is given by the ratio between the peak signal and the background measured off resonance. The resulting sensitivity is 500 ppb for CH₄. The background is calibrated replacing the CH₄ mixture with artificial air. The sensitivity is obtained without any post measurement treatment. Applications of techniques such as background subtraction should increase the sensitivity. Those measurements show that narrow linewidth nanosecond OPO are useful tools for spectroscopic applications at atmospheric pressure. The detectivity in the ppm range is appropriate for trace gas detection.



Figure 1 : Interference rings monitored with a 0.4 cm^{-1} free spectral range Fabry-Pérot. The OPO emission linewidth can be estimated to 0.08 cm^{-1} .

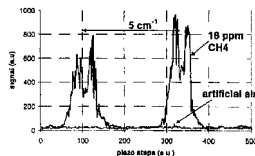


Figure 2 : CH₄ absorption line recorded by two successive resonances of the Fabry-Pérot. The 0.3 cm^{-1} double peak structure of the line is resolved.

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Laser Absorption Spectroscopy of Atmospheric Formaldehyde

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For the real time detection of atmospheric CH₂O with ultra high sensitivity and excellent selectivity, a cw tunable mid-IR spectroscopic source based on difference frequency generation (DFG) has been developed. The motivation for such a laser source is the acquisition of high quality CH₂O concentration measurements. Formaldehyde is an important intermediate present in all regions of the atmosphere and in testing the current understanding of hydrocarbon photochemistry. CH₂O plays also a critically important role in the chemistry of the urban atmosphere because of health effects associated with CH₂O, as well as its role in ground-level ozone formation.

The mid-infrared DFG source is based on low-power diode laser sources at 1 and 1.5 μm , which seed 1.5W Yb and 0.6W Er/Yb fiber amplifiers, respectively [1]. One seed source is a 1083 nm DFB diode laser, and 12 mW of its output was coupled into a single mode fiber to seed the Yb amplifier. A 2 mW fiber pigtailed 1.56 μm DFB telecommunications diode laser amplified to 30 mW with an Er fiber amplifier was used to saturate the gain of the Er/Yb fiber amplifier. A fiber beam coupler combines the two pump beams and delivers them to a lens for imaging the available pump power into a 19 mm long periodically poled lithium niobate (PPLN) crystal to produce up to 250 μW of cw DFG radiation in the PPLN crystal with grating periods ranging from 29.7 to 30.5 μm . The mid-infrared radiation is then collimated and directed into a 100 m long multipass absorption cell for CH₂O detection at reduced pressures. Various detection schemes were evaluated that included balanced detection with two MCT detectors, 2f-modulation spectroscopy, and CH₂O free zero air rapid background subtraction. The DFG wavelength is rapidly scanned using a sawtooth waveform ($\sim 0.17 \text{ cm}^{-1}$) across CH₂O absorption lines at 50 Hz.

This instrument was used both at NCAR in Boulder and at a rooftop location at Rice University in Houston, TX to acquire high quality CH₂O concentration measurement at the 1-20 ppbv level. At pressures of 40 torr, a temperature of 286 K and a pathlength of 100 m a minimum detectable absorbance of 10^{-3} was achieved with a measurement precision in the 0.2 to 0.3 ppbv range.

Reference:

[1] D.G. Lancaster, D. Richter, R.F. Curl, F.K. Tittel, L. Goldberg, J. Koplów, Opt. Lett. 24, 1744 (1999)