Modulation cancellation method for detection of molecules with unresolved absorption bands

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Abstract: A novel spectroscopic technique was applied to detection of hydrazine vapor using two wide stripe diode lasers and a variation of QEPAS. Modulating lasers with 180 degrees phase shift resulted in >100 times background reduction.

OCIS codes: (280.3420) Laser sensors; (140.5960) Semiconductor lasers

Vibrational spectra of most molecules consisting of more than five atoms are so dense that Doppler and pressure broadening make them unresolved at normal temperature and pressure conditions. As a result, infrared absorption spectra of such polyatomic molecules consist of almost unstructured bands, each 100-200 cm^{-1} wide. Spectroscopic identification of these species requires optical sources with wide spectral coverage, and the use of DFB or FP semiconductor lasers for this purpose is problematic.

In this work two independent wide stripe FP diode lasers were used to detect near-IR absorption of hydrazine (N_2H_4) vapor. Such lasers deliver high optical power that can exceed 1W, but usually are not used for spectroscopic applications because of high divergence and multimode nature (both spatially and spectrally) of the emitted radiation. It is not possible to efficiently couple radiation of a wide stripe laser into a multipass cell or an optical cavity. We solved this problem by applying a quartz tuning fork (QTF) to probe optical absorption in a gas via detection of acoustic waves generated in the gas upon absorption of modulated laser radiation. Thus, a version of quartz enhanced photoacoustic spectroscopy (QEPAS) technique was exploited [1]. In contrast to most QEPAS sensors, an acoustic microresonator was not used, thus reducing the probed optical path to the QTF thickness, 0.3 mm. The optical system created a magnified image of the 100 μm wide laser facet in the 0.3×0.3×3.6 mm^3 gas volume between the prongs of a QTF.

In practice, it is not possible to completely avoid irradiating the QTF by stray laser radiation, which results in unwanted background signal not related to optical absorption in the analyzed gas. A novel spectroscopic technique named the modulation cancellation method (MOCAM) was applied to suppress this background. The principle of this technique is illustrated by Fig. 1. In this figure, the laser 1 emission is spectrally positioned within the absorption band of molecule *A*, while laser 2 is detuned from this band. Radiation of the two lasers is combined and sent to the detector (a photoacoustic cell depicted in Fig. 1 as an example).

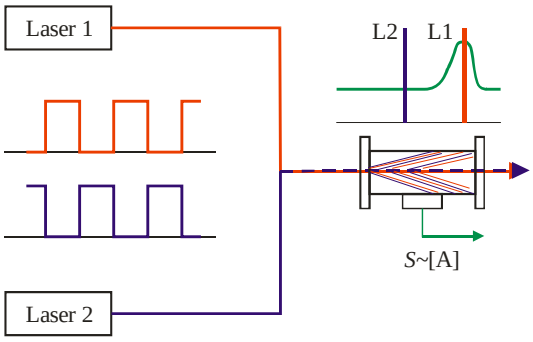

Fig. 1. Principle of MOCAM. Detected signal *S* is proportional to concentration of molecular species *A*.

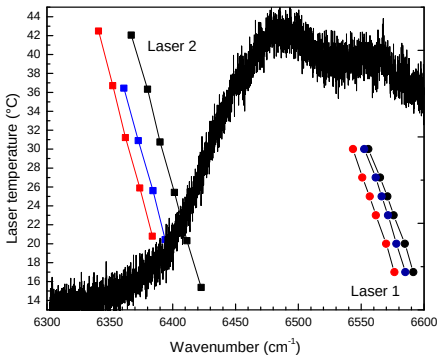

Fig. 2. Spectral position of the two lasers with respect to near-IR hydrazine absorption band (black line, PNNL database)

When the two lasers are modulated with a 180º phase shift and the intensities are balanced properly, the acoustic signal induced by spectrally nonselective absorption of stray radiation is cancelled, and the detected signal is proportional to spectrally selective optical absorption by chemical species *A*.

Spectral positions of the lasers used in this work are shown in Fig. 2 along with the hydrazine vapor absorption spectrum from the PNNL database. Different lines correspond to different laser injection currents in the 1 to 3 A range. The spectral width of each laser was ~40 cm^{-1}. An optical system used in the experiments is shown in Fig. 3.

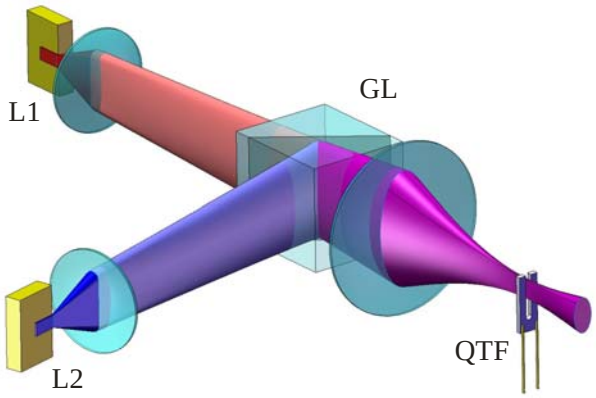

Fig. 3. Optical setup of the MOCAM-QEPAS based hydrazine sensor. L1, L2 – wide stripe lasers; GL – Glan prism; QTF – quartz tuning fork.

Both lasers emitted linearly polarized light. The polarization plane of one of the lasers was rotated 90º by means of half-wavelength plate (not shown in Fig. 3), and radiation emitted by the two lasers was combined using the Glan polarizer. The overlapped images of the laser facets were created between the QTF prongs. Injection currents of the lasers were modulated from threshold to an adjustable maximum value using sinusoidal waveforms with a 180º phase shift. Initial experiments showed an unwanted background suppression of 100 to 1000 times compared to unbalanced (one laser) detection. The hydrazine vapor detection limit was estimated to be ~1 ppmv.

References

[1] A. A. Kosterev, F.K. Tittel, D.V. Serebryakov, A. L. Malinovsky, I. V. Morozov, “Applications of quartz tuning forks in spectroscopic gas sensing,” Review of Scientific Instruments, 76, 043105 (2005).