TOWARDS OPTICAL DETECTION OF MOLECULAR SPECIES
AT SUB-PPT CONCENTRATION LEVELS

S. Bartalini, S. Borri, P. Cancio, I. Galli, G. Giusfredi, D. Mazzotti, P. De Natale
Istituto Nazionale di Ottica - CNR, Largo Fermi 6, 50125 Firenze FI
European Laboratory for Non-linear Spectroscopy, Via Carrara 1, 50019 Sesto Fiorentino FI

A frequency-comb-referenced cw difference-frequency-generated mid-IR source is combined with the saturated-absorption cavity ring-down spectroscopic technique to achieve optical detection of rare molecular isotopologues with sub-ppt concentration.

The fundamental (and strongest) ro-vibrational transitions of atmospheric molecules lie in the mid-IR region of the spectrum (the so-called "molecular fingerprint" region). Since trace-gas detection sensitivity scales inversely with line-strength, mid-IR is the best suited spectral region to perform high-sensitivity molecular spectroscopy.

With this motivation, we have designed and realized a difference-frequency-generated coherent source, with 3850-4540 nm tuning range and TEM\(_{00}\) spatial mode [1]. Phase locking of the near-IR "pump" laser to the "signal" one involved in the non-linear mixing process, through a Ti:sapphire-based optical frequency comb, also gives the generated "idler" radiation narrow linewidth and Cs-standard traceability [2]. The relatively high power of this source (30 mW at 4510 nm), greatly enhanced by a high-finesse Fabry-Perot cavity (F>11000), enables saturation spectroscopy at pressures even higher than 10 mbar.

By combining this source with our recently developed saturated-absorption cavity ring-down (SCAR) spectroscopic technique [3], we expect to achieve optical detection of rare molecular isotopologues with sub-ppt concentration. Indeed, the present spectrometer aims to detect partial pressures of the absorbing gas in the femtobar range per unitary noise bandwidth. More advantages of this setup are sub-Doppler resolution (at lower pressures), absolute frequency scale and absolute concentration measurement, with no need for calibrations.

The setup may also be further simplified by replacing the DFG source with a quantum-cascade laser (QCL), whose intrinsic linewidth was recently demonstrated to be much narrower than in conventional semiconductor lasers [4]. Nowadays, these compact, current driven mid-IR sources are commercially available with cw room-temperature operation. To achieve the frequency stability needed to be efficiently coupled to the high-finesse optical cavity employed in SCAR, a QCL can be easily frequency locked either to the cavity itself, or to a narrow molecular sub-Doppler line [5], by a current feedback loop.

Trace gas detection of molecular species can be crucial to get knowledge about physical, chemical, and biological processes. Many possible applications of such a compact and sensitive spectrometer span different everyday-life fields: medicine (absorption of drugs, disease diagnosis through breath analysis), environment (monitoring of air pollution), security (detection of hazardous substances, e.g. toxics and explosives).

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