A mid-IR QCL referenced to a fiber-based frequency comb applied to cavity ring-down spectroscopy for trace detection of CO_2 isotopologues

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Trace detection of CO_2, especially of the radioisotope ^{14}C is of high importance in radio-dating, as well as environmental and biological tracer applications, where isotope-labeled samples need to be investigated. We have previously demonstrated our capability to perform detection of ^{14}C using laser absorption based on cavity ring down spectroscopy (CRDS) at abundance levels lower than feasible with standard liquid scintillation counters [1,2].

To further improve our sensitivity and the reliability of the method many factors need to be considered, such as detector noise, temperature stability and etaloning effects. Additionally, at low abundance levels the isotope of interest has strong spectral overlap with absorption lines of contaminants and other isotopologues. In that case the precision of the frequency axis of the spectrum becomes vitally important to perform an accurate fit. Wavelength calibration of a mid-IR quantum cascade laser (QCL) was previously done via a solid silicon etalon as well as a N_2O absorption cell, reaching a frequency precision of the order of 10 MHz.

Frequency combs are well known as tools for precise optical frequency calibration but are not yet widely available in the mid-infrared region. A mid-IR fiber-based optical frequency comb was developed using a combination of a photonic crystal fiber and nonlinear crystal for single-pass difference frequency generation to the mid-infrared around 4.5 μm [3]. The output of the frequency comb and a fraction of the QCL light was overlapped in a single-mode fiber and sent to a fast thermoelectrically cooled photodetector after spectral filtering using a grating. The detected signal was analyzed with a fast spectrum analyzer and a beat-note signal with up to 20dB S/N ratio was observed.

Tracking as well as stabilization of the beat-note frequency was implemented in Labview via feedback to the QCL injection current and a frequency stability below 1 MHz was attained. Due to the low feedback bandwidth no laser linewidth reduction was possible. Nevertheless, even with other noise contributions in the ring-down spectra, the frequency calibration using the comb was found to provide higher reliability and repeatability of measured isotope ratios.

References

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