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QCL - Optical-Feedback Cavity Enhanced Absorption Spectroscopy For The Analysis Of Atmospheric $^{13}CO_2/^{12}CO_2$ In Ice-Core Gas Bubbles

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In the context of a globally warming climate it is crucial to study the climate variability in the past and to understand the underlying mechanisms. The composition of gas stored in bubbles in polar ice presents a paleo-climate archive that provides a powerful means to study the exact mechanisms involved in the ∼40% increase in the atmospheric CO_2 concentration between glacial and interglacial climates. It is particularly important to understand such natural coupling between climate and the carbon cycle, as it will partly determine what natural feedback can be expected on the atmospheric CO_2 concentration in a future warmer world. The source of the CO_2 released into the atmosphere during previous deglaciations can be constrained from isotopic measurements by the fact that the different CO_2 reservoirs (terrestrial biosphere, oceans) and associated mechanisms (biological or physical) have different isotopic signatures. Unfortunately, such isotope studies have been seriously hampered by the experimental difficulty of extracting the CO_2 without contamination or fractionation, and measuring the isotope signal off-line on an isotope ratio mass spectrometer (IRMS).
Here we present an alternative method that leverages the extreme sensitivity afforded by Optical Feedback Cavity Enhanced Absorption Spectroscopy (OF-CEAS) in the Mid-Infrared [1]. This region of the spectrum is accessed by a custom-developed Quantum Cascade Laser operating near 4.35 μm. The feedback to the laser of light that has been spectrally filtered by a high-finesse, V-shaped enhancement cavity has the effect of spectrally narrowing the laser emission and to auto-lock the laser frequency to one of the cavity's longitudinal modes, with clear advantages in terms of acquisition time and signal-to-noise ratio of the measurement. The line strengths in this region are about 5 orders of magnitude higher than in the more easily accessible NIR region near 1.6 μm and about 1000 times higher than at 2 μm. The instrument is temperature stabilization at the mK-level. Together with a small cavity volume of ∼20 mL, this enables the analysis of nmol-sized samples with high precision (< 0.05‰ in a fraction of the time required by the conventional IRMS-based technique. We will show preliminary results obtained on synthetic samples.

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[2] Morville J., Kassi S., Chenevier M., and Romanini D.: Fast, low-noise, mode-by-mode, cavity-enhanced absorption spectroscopy by diode-laser self-locking, Appl. Phys., B 80, 1027-1038, 2005.