Doppler-Broadened Mid-Infrared NICE-OHMS System Based on an Optical Parametric Oscillator

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Although detection sensitivities in the 10^{-14} cm^{-1} range recently has been demonstrated by fiber laser based Doppler broadened (Db) NICE-OHMS in the near-infrared (NIR) region, it has so far not been possible to utilize the full power of the technique for trace gas detection since molecular species in general only have weak overtone bands in this wavelength region. The realization of a Db NICE-OHMS instrumentation working in the mid-IR (MIR) region, in which many molecules have strong fundamental vibrational bands, is however not straightforward due to the lack of electro-optic modulators (EOMs) in this wavelength range. To circumvent such problems, we have realized a MIR NICE-OHMS system around a resonant optical parametric oscillator (OPO) pumped by a narrow linewidth fiber laser lasing in the 1064 nm region. By modulating the pump laser light by an ordinary EOM, and letting the signal wave be locked to a cavity mode of the OPO, a frequency modulated idler output in the 3.2-3.9 μm region results. This opens up for NICE-OHMS detection of not only trace of gases to low concentrations, but also their isotopologues. The laser was first locked to a low finesse cavity (F = 500). An Allan-Werle analysis revealed a white noise equivalent absorption per unit length (NEAL) of 3×10^{-9} cm^{-1} $Hz^{-1/2}$ and a minimum NEAL of 1.5×10^{-9} cm^{-1} for a 20 s integration time, which corresponds to 90 ppt of CH4 detected at 40 Torr. Further improvement of this system, e.g. by improving the locking by the use of an acousto-optic modulator (AOM) so that a higher finesse cavity (F = 5000) could be implemented, have since then been pursued, and we are presently approaching a NEAL of 1×10^{-10} cm^{-1}. This opens up for a number of interesting applications. Since the origin of the methane in nature can be assessed by the relative concentration of its isotopologues, we aim, as a first application, for detection of $^{13}CH_4$ and CH_3D, which exist in ppb and sub-ppb concentrations, with good accuracy.