High-Accuracy Molecular Spectroscopy with Frequency Comb-Linked Mid-Infrared Continuous-Wave Optical Parametric Oscillator

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The mid-infrared (MIR) region contains strong fundamental ro-vibrational transitions (for example, CH, NH, and OH stretching modes) of various molecules. The strong absorption and the large number of detectable molecules make this region ideal for spectroscopy. The accuracy and the usefulness of spectroscopic techniques rely on the quality of spectroscopic databases, such as HITRAN [1]. These databases list absorption line parameters, such as line strength, center frequency, width, and line broadening coefficients for several molecular species and isotopologies. Using the listed parameters it is possible to simulate molecular absorption spectra, which helps to find the optimum spectral region for spectroscopic detection of a given molecular species. The simulations are essential for data retrieval, which is based on fitting of the simulated spectra to the measured absorption spectrum. Measurement accuracy in trace gas analysis therefore directly depends on the quality of spectroscopic data in the databases, and severe errors can result from inaccuracy of such data [2].

We report here an accurate mid-infrared laser spectrometer that has been designed to measure molecular spectroscopic data, in order to improve the quality of the existing spectroscopic databases. The spectrometer is based on a wavelength tunable cw OPO [3], which is used for high resolution cavity ring-down spectroscopy (CRDS) between 3 and 4 µm mid-infrared wavelengths. The absolute frequency of the idler beam (which is used for spectroscopy in the MIR) of the cw OPO can be determined by measuring the pump and signal frequencies using an optical frequency comb (OFC) [4-6]. Another possibility, which is used in this work, is to frequency double the MIR idler beam, after which its frequency can be referenced to an OFC. The principle of this approach is illustrated in Fig.1. In our conference contribution, we will present a detailed description of the spectrometer, as well as absolutefrequency measurements of specific rotational lines of the R branch of the v_3 vibrational band of CH_4.

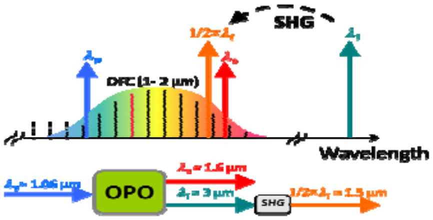

Fig. 1. The principle of absolute frequency measurement with the MIR cw OPO. The cw OPO is pumped at λ_p = 1064 nm and it is singly resonant at λ_s ~1.5 µm. The absolute frequency of the idler beam (λ_i~3 µm) is measured with a frequency comb after frequency doubling (SHG) in a nonlinear crystal.

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