High-precision spectroscopic system based on a frequency-comb-assisted quantum cascade laser at around 8.6 µm

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We report on metrological-grade spectroscopic system based on a distributed-feedback quantum cascade laser (QCL), tunable in the wavelength range from 8.56 to 8.63 µm, phase-locked to a tooth of a difference frequency generation (DFG) mid-IR frequency comb [1]. The beat note is directly generated in the mid-IR region without any conversion scheme resulting in a compact system that combines the broad-band parallel detection capabilities of a mW-level mid-IR comb to the high-precision performances of a mid-IR narrow linewidth source. The obtained ~30 dB signal-to-noise ratio of the detected beat-note together with the achieved closed-loop locking bandwidth of ~500 kHz allows for a residual integrated phase noise of 0.78 rad (1 Hz - 5 MHz), for an ultimate resolution of ~21 kHz, limited by the measured linewidth of the mid-IR comb. By tuning the comb repetition rate, the QCL frequency is scanned with an ultimate accuracy of a GPS-disciplined Rb frequency standard. This system is a powerful tool for high-resolution molecular spectroscopy in the mid-IR spectral region. In particular, it will be used for the absolute measurement of the line center frequency of a two-photon transition in a molecular beam of CHF_3 allowing for a determination of the possible variation of the electron to proton mass ratio at level of 10^{-15}yr^{-1} [2].

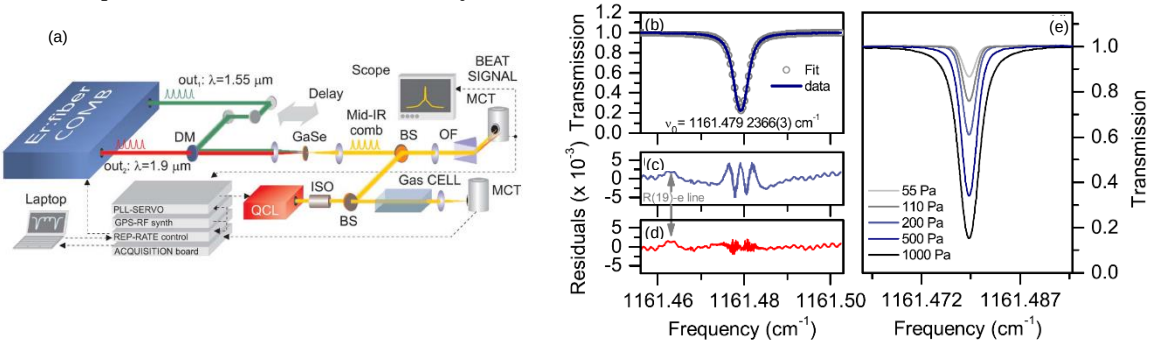

Fig. 1 (a) Experimental setup for the comb-assisted QCL at 8.6 µm. (b) Averaged transmission profile (five scans) of the P(8)-e rotovibrational line of N_2O. Residuals of the (c) Voigt and (d) Galatry fits. (e) Transmission profile at different N_2O pressures.

Figure 1 (a) shows the experimental setup for the DFG comb synthesis (based on a femtosecond Er fiber comb [1]) and the QCL phase-locking. Preliminary spectroscopic measurements of the ν_2 vibrational band of N_2O molecule have been realized to check the system performance. We performed several acquisitions of the N_2O P(8)-e rovibrational transition at different pressure in the range from 55 to 1000 Pa (at a temperature of 296 K), as shown in fig. 1 (b) and 1 (e). A linear fit of the retrieved line-centre frequencies as a function of the gas pressure allowed us to extrapolate a zero-pressure value of the center frequency of 1161.4792366(3) cm^{-1} (corresponding to a relative precision of 3×10^{-10}). This value deviates from the one of the HITRAN database by 2.6×10^{-5} cm^{-1}, well within the HITRAN accuracy, and results to be more than two orders of magnitude more precise. The statistical uncertainty on the extrapolated zero-pressure value of the line-center frequency is mostly limited by the SNR of the recorded spectra and by the pressure-dependent repeatability of the line-center frequency retrievals. On the other hand, the main source of systematical deviation is the pressure reading (whose contribute amounts to 6×10^{-10}); other sources, including the electronic detection chain bandwidth and the RF-standard accuracy are negligible, being at the 2×10^{-12} level. Further experimental investigations on both N_2O and CF_3H molecules are in progress and the detailed spectroscopic results will be presented during the CLEO Europe 2015 conference.

References

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