

A Widely Tunable 10 μm QCL Locked to a Metrological Mid-IR Reference for Precision Molecular Spectroscopy

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The quantum cascade lasers (QCL) are popular sources for spectroscopy in the field of mid-infrared because of the wide range of wavelengths they can cover ($3 \mu\text{m} < \lambda < 24 \mu\text{m}$) [1]. Several examples of spectroscopic measurements with spectrometers based on QCL have been reported [2].

We are currently developing a laser spectrometer based on a near-room-temperature distributed feedback QCL which emits around $10 \mu\text{m}$. The QCL source has been compared to an ultra-stable CO_2 laser.

We characterized the free-running QCL and demonstrated a frequency noise roughly one order of magnitude lower than previously published characterizations of QCLs. A full width at half maximum (FWHM) of 60 kHz for the beat signal between the free-running QCL and a 1-kHz narrow CO_2 laser was observed for 1 ms of integration time.

We have also demonstrated a narrowing of the QCL by a coherent phase-lock to a CO_2 laser stabilized onto a saturated absorption line of OsO_4 . The beat spectrum (see Fig. 1) between phase-locked QCL and CO_2 laser recorded with a radio-frequency (RF) spectrum analyzer allowed us to estimate that more of 99% of the beat signal RF power is concentrated in the laser carrier. This allows to conclude that the QCL reproduces almost exactly the spectral characteristics of our ultra-stable CO_2 laser (10-Hz line width, accuracy of a few tens of hertz). This results in record QCL line width of the order of 10 Hz, 3 to 4 orders of magnitude lower than a free-running QCL, and a relative stability at 1 s of about 1 Hz.

The phase-locked QCL was then used to record spectra of ammonia (NH_3) and methyltrioxorhenium (MTO) (see Fig. 2) to demonstrate its potential for two main projects of our group: the determination of the Boltzmann constant, k_B , by Doppler spectroscopy of ammonia [3] and the first observation of parity violation in chiral molecules [4].

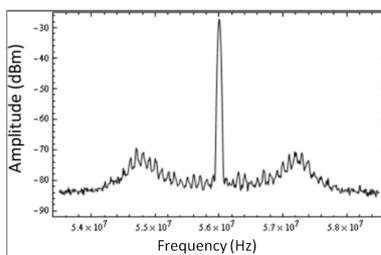


Figure 1: Beat signal spectrum between the frequency-stabilized CO_2 laser and the phase-locked QCL recorded with a RF spectrum analyzer (30 kHz resolution band-width).

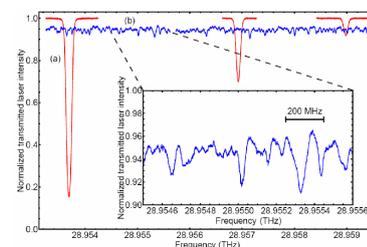


Figure 2: Linear absorption spectra of NH_3 (red curve (a)) and MTO (blue curve (b))

References

- [1] Kosterev *et al.*. Applied Physics B, **90**(2), 165-176 (2008)
- [2] Wysocki *et al.*. Applied Physics B, **92**(3), 305-311 (2008)
- [3] C. Lemarchand *et al.* Metrologia **50** 623 (2013)
- [4] S. K. Tokunaga *et al.* Molecular Physics 111, Issue 14-15, 2363–2373 (2013)