

## Subkilohertz-narrowed, frequency/phase-locked mid-IR quantum cascade lasers for high-precision molecular spectroscopy

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The interest in the development of narrow-linewidth mid-IR sources is related to advanced applications such as trace-gas sensing with high-finesse cavities, high-precision spectroscopy and frequency metrology. For all these fields, the mid-IR portion of the electromagnetic spectrum, named also "molecular fingerprint region", is the best suited for investigating many molecules of atmospheric and astrophysical interest, whose strongest ro-vibrational transitions lie in this spectral region.

Our group has recently demonstrated that the spectrum of the radiation emitted by a quantum cascade laser (QCL) can be narrowed through the optical injection technique [1]. We show that at least two more techniques can be used to narrow the spectrum of the radiation emitted by a QCL down to a subkilohertz level. The first one involves the frequency lock to a molecular absorption line detected by polarization spectroscopy [2]. This method permits to produce a dispersive signal that can be directly processed by a PID controller for correcting the laser frequency fluctuations. With this experiment we have demonstrated the possibility to reduce the QCL emission linewidth from 500 kHz down to less than 1 kHz, a value close to the laser intrinsic linewidth. The same narrowing performance can be achieved by the second technique, relying on phase locking the QCL to a narrow-linewidth radiation generated by a difference-frequency process (DFG) in a periodically-poled LiNbO<sub>3</sub> non-linear crystal [3]. This DFG source is absolutely referenced to the Cs primary frequency

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standard through a visible/near-IR optical frequency-comb synthesizer (OFCS), acting as a transfer oscillator across the 70-THz gap between the "pump" and "signal" lasers generating the "idler" mid-IR radiation.

The importance in narrowing and absolutely referencing the radiation emitted by QCLs lies in the possibility to perform spectroscopic analysis with increasing resolution, accuracy and sensitivity. In fact, QCLs are nowadays the most compact electrically driven devices capable of emitting coherent radiation in mid and far IR with appreciable power (from mW to W). The phase-locking technique has been already applied to saturated-absorption Lamb-dip spectroscopy of CO<sub>2</sub> ro-vibrational transitions at 4.3 μm, whose line-center frequencies have been measured with few kHz uncertainty [4]. We have also proposed the frequency-locking technique of the QCL for replacing a complex OFCS-referenced intracavity DFG source used for optical radiocarbon detection based on saturated-absorption cavity ring-down (SCAR) with a much simpler and compact source [5]. The subkilohertz narrowing is crucial for coupling the QCL radiation to a high-finesse cavity with about 10 kHz resonance width, while the frequency lock to a molecular transition further simplifies the SCAR experimental setup, removing any need for the OFCS reference.

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[3] I. Galli, M. Siciliani de Cumis, F. Cappelli, S. Bartalini, D. Mazzotti, S. Borri, A. Montori, N. Akikusa, M. Yamanishi, G. Giusfredi, P. Cancio, and P. De Natale, *App. Phys. Lett.* 102, 121117 (2013).

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[5] I. Galli, S. Bartalini, P. Cancio, P. De Natale, D. Mazzotti, G. Giusfredi, M. E. Fedi, and P. A. Mandò, *Radiocarbon* 55 (2013).