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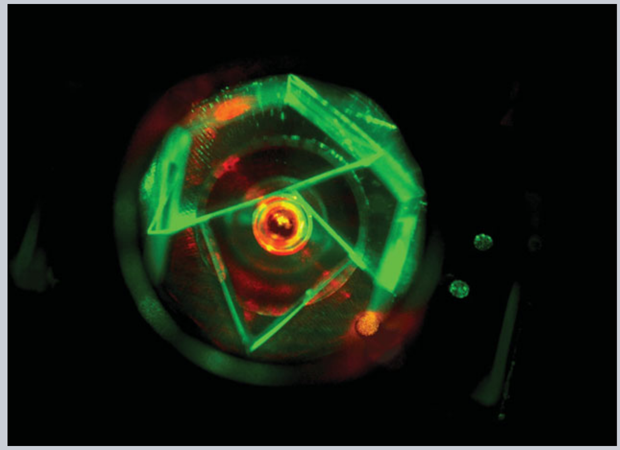
Narrow-linewidth lasers are key elements in optical metrology and spectroscopy. Spectral purity of these lasers determines accuracy of the measurements and quality of collected data. Solid state and fiber lasers are stabilized to relatively large and complex external optical cavities or narrow atomic and molecular transitions to improve their spectral purity. While this stabilization technique is rather generic, its complexity increases tremendously moving to longer wavelenghts, to the infrared (IR) range. Inherent increase of losses of optical materials at longer wavelengths hinders realization of compact, room temperature, high finesse IR cavities suitable for laser stabilization. In this paper, we report on demonstration of quantum cascade lasers stabilized to high-Q crystalline mid-IR microcavities. The lasers operating at room temperature in the 4.3-4.6 μm region have a linewidth approaching 10 kHz and are promising for on-chip mid-IR and IR spectrometers.



LETTER ARTICLE

Microcavity-Stabilized Quantum Cascade Laser

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The quest for tunable and spectrally pure laser sources in the mid-infrared (mid-IR) is one of the most challenging photonic endeavours of the last decades. High-sensitivity spectroscopic techniques based on high-finesse optical resonators have proved to greatly benefit from narrow-linewidth lasers, achieving record sensitivity values for trace gas sensing [1]. Highly-integrated, compact and frequency-stabilized laser sources having mid-IR emission wavelengths are needed to replace conventional bulky spectroscopy designs for fieldable mid-IR spectrum analyzers. The lasers must have high spectral purity, minimal sensitivity to optical feedback, long lifetime, reliable wavelength tuning and setpoint repeatability to ensure high accuracy and reliable measurements. Furthermore, due to the Hz-level natural linewidths for vibrational and rotational molecular transitions in the IR, there is a growing interest even for high-precision spectroscopy of molecules useful to test fundamental physics, e.g. for measuring fundamental constants and their possible time variation [2–4] or testing fundamental symmetries [5]. Quantum Cascade Lasers (QCLs) are promising sources for these applications because of their broad spectral coverage and ultranarrow fundamental linewidths [6]. Such sources, though, suffer from instrumental excess noise, occurring due to temperature drifts and electrical noise. A proper stabilization technique must be implemented to reduce the technical noise in order to fully exploit the inherent advantages of QCLs.

Several techniques have been devised to narrow the linewidth of mid-IR QCLs. Phase-locking to infrared frequency combs [7] or frequency-stabilization over sub-Doppler molecular transitions [8] allowed achieving sub-kHz linewidths. However, in the former case an optical comb is required, while the second approach is viable only when a suitable molecular transition is available. QCLs locked to stable mid-IR references obtained via coherent frequency down-conversion demonstrated sub-10 Hz absolute linewidth [9]. This approach, though, is intended for well developed metrology laboratories and is too complex for field applications. The same problem is related to QCL stabilization with optical super-stable cavities. This method has been successfully implemented and allowed achieving a linewidth as low as 60 Hz [10], but required upconversion of mid-IR frequency to the near-IR region in a bulky setup.

Highly-integrated and compact approaches are needed to replace expensive and bulky stabilized mid-IR sources, and direct lock of QCLs to mid-IR cavities stands out as the most attractive solution. It was shown that optical feedback (self-injection) locking to a high-finesse V-shaped cavity narrows the 1 ms QCL linewidth from 3.2 MHz to 4 kHz FWHM [11]. However, this configuration sufferes from

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the use of a large, non-monolithic cavity which can be prone to environmental noise. Indeed, despite the noticeable narrowing at short timescales, the QCL linewidth remained as broad as several MHz over longer timescale (>1 s).

Creating a chip-scale Fabry-Perot cavity for self-injection stabilization of a QCL is not straightforward. A large cavity size allows achieving the high quality (Q-) factor needed for linewidth reduction. Compact cavities require better mirrors to achieve similar Q. This is not trivial, since materials used for mirrors are usually lossy at longer wavelength. The use of monolithic mid-IR whispering-gallery mode resonators (WGMRs) offers a solution to this problem. The combined miniaturization of the laser/resonator systems, achievable by using QCLs and WGMRs, is attractive for various applications, as was very recently shown by mid-IR Kerr frequency comb generation [12–14].

In this work, we demonstrate QCLs frequency stabilization and linewidth narrowing by locking to high-Q ($>10^7$) mid-IR WGMRs. This method, validated at optical wavelengths [15], has multiple salient advantages in mid-IR. Differently from standard Fabry-Perot cavities, whose highly reflective mirrors operate at very specific wavelengths, our resonators work over a wide spectral range, from below 1 μm to over 5 μm wavelength, and can be used for stabilization of very different lasers. While it is difficult to increase the finesse of standard optical resonators (due to the difficulty of making highly reflecting mirrors, especially at long wavelengths), mid-IR crystalline resonators are at the very beginning of their story and their Q-factors are expected to increase with the improvement of materials as well as of fabrication techniques, thus offering a wider applicability perspective than standard cavities. Microcavities, like those used in this work, are compact and highly versatile, lending themselves for monolithic integration and development of on-chip IR spectrometers. We implemented two complementary methods for QCL stabilization, using self-injection locking as well as all-electronic locking onto the transmission mode, thus demonstrating record stability of QCL operation over the entire spectral noise frequency range. We achieved ≈10 kHz linewidth (full width at half maximum, FWHM) over a 1 s timescale, an unprecedent results for direct locking to a mid-IR optical resonator.

Electronic locking Our first method, electronic stabilization of QCL to the transmission mode of the resonator, was selected for the robustness of the technique. The experiment, performed at the INO labs in Firenze, was based on the setup schematically shown in Fig. 1. A room-temperature single-mode distributed-feedback (DFB) continuous wave QCL operating at 4.3 μm wavelength (from Hamamatsu Photonics) was mounted in a home-made housing provided with thermoelectric cooler and collimating aspheric lens. The laser, driven by a homemade ultra-low noise current driver, was equipped with a modulation input (DC-1 MHz bandwidth), and a bias-tee allowed applying fast modulations (exceeding tens of MHz) directly to the laser chip. The QCL had 700 mA threshold current at 288 K temperature. The laser delivered 10 mW output power at 780 mA current.

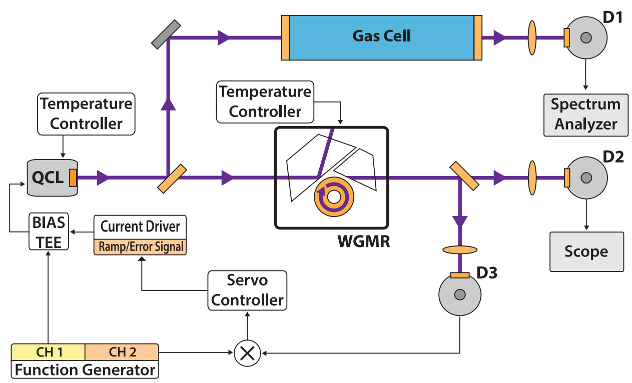

Figure 1 Schematic of the setup used for electronic frequency stabilization. D1 and D2: InSb photodiodes; D3: Vigo detector.

A CaF_2 toroidal WGMR from OEwaves was used for electronic stabilization of the QCL. The resonator had a diameter of 3.6 mm, corresponding to a free-spectral range of 18.9 GHz at the experimental wavelength, and was mounted inside a custom-made housing in order to reduce both thermal and mechanical fluctuations, and to protect it from dust and humidity. The QCL was free-beam coupled to the resonator through a coupling prism, placed close to the resonator surface (prism-WGMR gap $\simeq$0.5 μm at the working temperature of 307.5 K). Optimal coupling required beam waist of about 10 μm (radius at $1/e^2$ of the total beam power). In operating conditions, the measured WGMR transmission mode was 3.1 MHz FWHM, corresponding to a Q$\simeq$2.2·10^7. Thanks to the large distance between the laser and the resonator, and to a few irises along the path, no significant feedback was measured onto the laser due to backscattered radiation.

About one half of the power exiting the laser was sent to a spectroscopic cell filled with pure CO_2 and was detected by a liquid N_2 cooled InSb photodiode. The detected signal was acquired by a real-time spectrum analyzer (Tektronix RSA 3303A, DC-3 GHz band). The radiation transmitted through the resonator was splitted into two parts by means of a beam-splitter. About 25% of the radiation was sent to another liquid N_2 cooled InSb photodiode which monitors the signal level in locked and unlocked conditions. The remaining 75% was detected by a termoelectrically cooled fast photodiode (Vigo PVI-4TE-5, ~10 MHz bandwidth), and the signal was processed by the locking electronics (frequency mixer and servo controller) and added to the laser current as a correction signal via the modulation input.

A waveform generator was used to add a slow triangular ramp (100 ms period) to the laser by the modulation input of the current driver to scan the frequency through the resonator modes. In order to generate an error signal, a fast sinusoidal modulation was added to the laser frequency by means of a double-channel signal generator. One channel was used to couple the sinusoidal modulation to the laser pad through the bias-tee. The second output channel, which allowed generating a sinusoidal signal with the same frequency as the first one but with a different phase offset, was

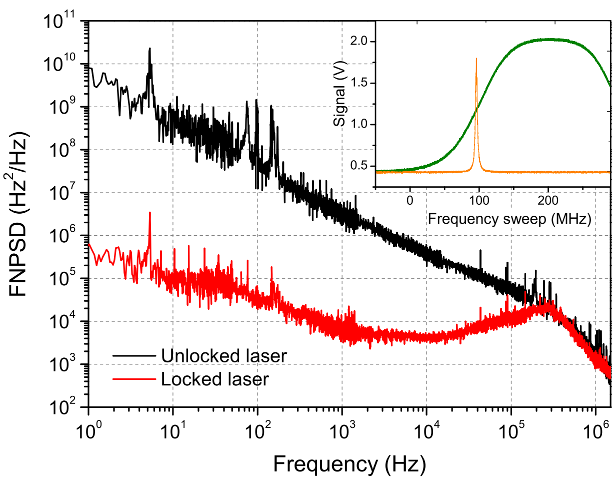

Figure 2 Frequency-noise power spectral density for free-running and locked (electronic stabilization) laser (black and red traces, respectively). In unlocked conditions flicker noise dominates up to the detector cut-off (∼300 kHz). Inset: operating conditions for frequency noise measurements: the WGMR locking transmission mode (orange peak) is placed at the center of the slope of a strong CO_2 absorption line (green curve, 1 mbar pressure).

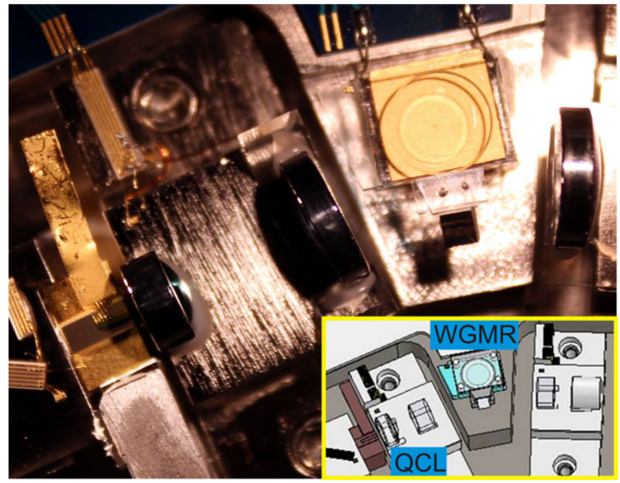

Figure 3 A picture and schematic (inset) of the experimental setup used for self-injection locking of a DFB QCL to a WGMR. Light from a QCL was collimated using Si lens and coupled to a CaF_2 WGMR via a BaF_2 evanescent field prism coupler. The light exiting the resonator was collimated and processed. The locking occurred due to resonant Rayleigh backscattering in the WGMR.

used as local oscillator for the mixer. The servo controller consisted of two independent stages, a proportional and an integral one, and allowed to exclude the slow ramp in locking conditions. By switching on the fast modulation on the laser, the error signal was generated at the mixer output. We selected optimal values for the laser fast modulation (3.5 MHz frequency and ≃5 MHz modulation depth) by direct observation of the error signal.

In order to compare the frequency fluctuations of the laser in free-running and locked conditions, we measured its frequency-noise power spectral density (FNPSD) by using the side of a strong molecular absorption line as frequency-to-amplitude converter. We selected the strongest CO_2 line observable within the tuning range of the laser, the (000-001) P(42) transition occurring at 2311.105 cm^{-1} (4.327 μm), with a linestrength of $4.75 \cdot 10^{-19}$ cm (HITRAN units). We choose the CO_2 pressure inside the spectroscopic cell in order to maximize the slope of the absorption line (P≃1 mbar) for an optimal frequency-to-amplitude conversion. The microresonator temperature was chosen in order to have one of its modes centered on the side of the absorption line (see inset in Fig. 2). In such conditions, we recorded the spectrum of the transmitted intensity, which reproduces the spectrum of the laser frequency fluctuations scaled by the slope of the absorption profile.

The measured laser FNPSD is shown in Fig. 2. The locking bandwidth is about 250 kHz, limited by the laser frequency response [16], and the loop is able to pull down the laser FNPSD by more than 3 orders of magnitude, far below 1 kHz/$Hz^{1/2}$. By using the technique proposed in [17] we inferred ≃700 kHz linewidth for the free-running laser (1 s timescale), which is reduced to 15 kHz in the locked regime (10 kHz for 1 ms timescale). The stability at long timescales is impressive, and marks the difference with QCL locking to mid-IR Fabry-Perot cavities [11]. In high-resolution spectroscopic measurements this may allow for long averaging times, even if the WGMR is not stabilized to any frequency reference (a residual excess noise is visible in the low frequency part of the spectrum).

Self-injection locking The second technique for QCL stabilization explored in our study is based on optical self-injection locking. The laser stabilization quality, in this case, depends on integration of a QCL chip and a WGMR. Both the laser and the resonator have to be integrated on the same optical micro-bench in order to avoid mechanical instabilities along the optical path. Preliminary tests of this technique were performed in the OEwaves labs in Pasadena using 4.55 μm QCL on C-mount (Adtech) and a 6 mm diameter CaF_2 WGMR characterized with 3 MHz bandwidth at this wavelength.

The setup is shown in Fig. 3. The resonator was integrated with evanescent field coupler and the DFB QCL chip and placed on a thermally stabilized optical micro-bench. There was no isolator between the laser and the resonator. Part of the light was backscattered to the laser chip when the laser frequency coincided with a WGM, resulting in the self-injection locking effect.

The mid-IR light emitted by the laser was sent to a photodiode through an isolator as well as a frequency discriminating mid-IR optical WGM filter with approximately 20 MHz bandwidth. Again, as the spectrum of the laser intensity noise was found to be much smaller than that of the signal transmitted by the filter, from the latter the phase noise of the laser could be retrieved. We performed the experiment for locked and unlocked QCL and found that the frequency noise of the laser drops at least by 40 dB when measured at 10 kHz offset frequency.

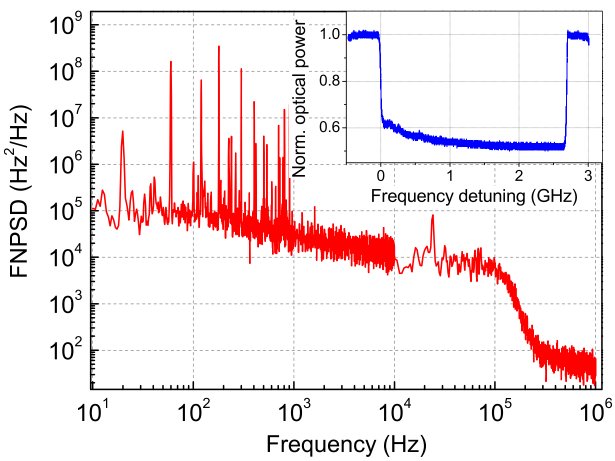

Figure 4 Frequency-noise power spectral density of the self-injection locked DFB QCL. Inset: Dependence of the power of the mid-IR light leaving the resonator on the frequency detuning between the partial frequency of the free running QCL and the resonator mode the laser is locked to.

The measured FNPSD of the self-injection locked QCL is shown in Fig. 4. The dynamic range of the measurement was limited because of the technique used. At small frequencies we were unable to perform measurements when the corresponding phase noise exceeded 0 dBc/Hz (free-running conditions). At high frequencies the measurement accuracy was limited by our photodiode response (frequency noise cut-off above 100 kHz shown in the graph).

Inset of Fig. 4 shows the dynamic range of the self-injection locked laser. The area of reduced output power corresponds to sustained locking conditions. The locking range was measured by changing the QCL current via the sweepable Wavelength Electronics low-noise laser driver. The output of the laser-WGMR system was sent trough an Innovation Photonics isolator and collected with a mid-IR fiber with gold-coated focusing mirror. The fiber output was connected to a PDA10JT HgCdTe photodiode, and the detected signal was analyzed using a Tectronix oscilloscope. The QCL frequency tuning range was calibrated to be 670 MHz/mA. We detected a self-injection locking range of ∼5 mA corresponding to 3.35 GHz in frequency units.

It is interesting to compare the FNPSD of the electronic and self-injection locked lasers (see Fig. 2 and Fig. 4). The two measured frequency noise dependencies show similar trends and values, with a slight excess noise in the low-frequency part of the spectrum of the self-injection locked device and an almost flat plateau above a few kHz. This is remarkable, as the setups are completely different. They coincide only in bandwidths of the WGMRs used for QCL stabilization. The results of our experiments confirm that it is feasible to obtain QCLs with less than 10 kHz linewidth using WGMR resonators. While the measured limitations of the frequency noise can be attributed to WGMR properties, a more detailed study is required to understand fundamental limitations of the explored stabilization techniques when applied to room temperature DFB QCLs.

In conclusion, 10-kHz level mid-IR QCLs have been demonstrated by locking to high-Q crystalline microresonators. This approach allows for narrow-linewidth tunable lasers with a compact and robust package, paving the way for the realization of very compact spectrally pure mid-IR spectrometers suitable for high-resolution molecular sensing. It is worth noting that the achieved long-term stability is particularly attractive for demanding spectroscopic measurements, as it allows for effective averaging over time without loosing in spectral resolution.

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