Subkilohertz linewidth room-temperature mid-infrared quantum cascade laser using a molecular sub-Doppler reference

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We report on the narrowing of a room-temperature mid-IR quantum cascade laser by frequency locking it to a CO_2 sub-Doppler transition obtained by polarization spectroscopy. A locking bandwidth of 250 kHz has been achieved. The laser linewidth is narrowed by more than two orders of magnitude below 1 kHz, and its absolute frequency is stabilized at the same level.

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The demand of single-frequency, high-power, narrow-linewidth laser sources is soaring for applications ranging from high-sensitivity spectroscopy to frequency metrology, environmental sensing, cold atoms, and molecule control. Thanks to the development of optical frequency comb synthesizers (OFCSs), this quest has, so far, led to impressive results mainly in the visible and near-IR spectral regions, yielding a large number of successful metrological applications.

Moving to longer wavelengths, comb-referenced mid-IR sources based on frequency mixing in nonlinear crystals [1] have been demonstrated to be highly performing for high-precision [2,3] and high-sensitivity spectroscopy [4], especially when coupled to high-finesse resonators [5]. Together with absolute-frequency referencing, these sources achieve linewidths as narrow as a few kilohertz, as required for an efficient coupling to the narrow modes of the high-finesse cavity.

The rapid development of mid-IR quantum cascade lasers (QCLs) over the last decade has made available compact and versatile sources, with intrinsic linewidths of only a few hundred hertz [6]. Moreover, a recent study confirmed that the QCL's emission spectrum gets narrower when moving from cryogenic to room-temperature (RT) operation [7]. On the other hand, RT operation may produce other effects on key parameters: by comparing the current-to-frequency tuning rates of an RT [8] and a cryogenic [9] QCL, a degradation of the tuning capability at higher temperatures can be inferred.

In this framework, a crucial step toward an extensive use of QCLs for demanding spectroscopic and metrological applications is the development of techniques enabling not only the narrowing of the QCL emission down to the kilohertz level but also its referencing to a stable frequency standard.

Up to now these two goals have been pursued separately. Frequency stabilization of mid-IR QCLs to stabilized high-finesse optical cavities [10] or to the side of a Doppler-broadened molecular transition [11] achieved narrowing of the QCL emission but no absolute frequency referencing. On the other hand, frequency locking of a QCL on the center of a sub-Doppler molecular line [12] or phase locking to an OFCS [13] provided an absolute reference of the QCL frequency, but did not achieve any significant narrowing of its emission. A powerful approach to providing a narrow and absolutely-referenced QCL emission can be phase-locking the QCL to an ultra-narrow OFCS-based source. This last option will probably be the best solution for the most demanding applications, though at the price of a greater complexity and cost.

In this Letter, we present an approach to a narrow-emission, absolutely referenced mid-IR QCL. It exploits the availability of a natural ruler of frequency references given by the many strong molecular absorption lines, whose center frequency can be absolutely measured with a subkilohertz precision [14]. Based on this, it is possible to have a simple system for high-sensitivity/precision spectroscopy for a specific molecular species, without using an OFCS. A polarization spectroscopy (PS) scheme [15] produces, without any external modulation, the narrow sub-Doppler signal used to close the feedback loop on the QCL driving current for frequency stabilization. We show that the linewidth of a cw RT QCL can be narrowed below 1 kHz (FWHM) by locking the laser to a CO_2 line.

The laser is an RT distributed feedback (DFB) QCL emitting at 4.3 μm provided by Hamamatsu Photonics, from the same processing of the QCL we characterized in [16]. It is operated at a temperature of 283 K and a current of 710 mA, delivering an output power of about 10 mW. A schematic of the experiment is shown in Fig. 1. The spectroscopic setup is similar to that described in our previous work [15], with some significant changes that strongly improve the stability of the PS signal. In particular, the use of a compact thermoelectrically cooled laser mount, instead of a liquid-N_2 cryostat, provides a dramatic

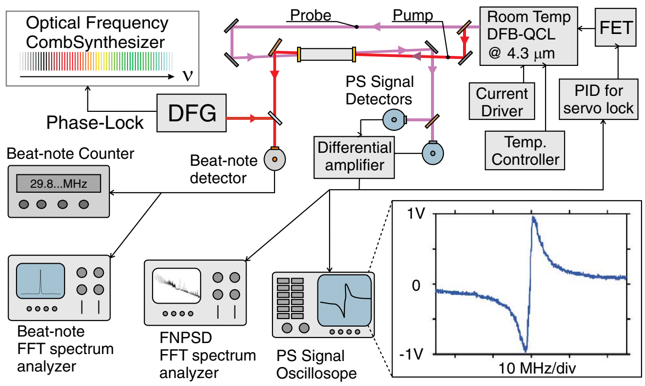

Fig. 1. (Color online) PS setup (for details, see [15]). The "probe" beam gives the signal used for the frequency lock. The "pump" beam is also used for the beat-note detection and the frequency counting. DFG, difference frequency generator; FFT, fast Fourier transform.

reduction in the pointing instabilities of the laser beam. Significant improvements have been obtained also on the noise performance of our home-made current driver. It now ensures a current-noise power spectral density that is always below 1 $\mathrm{nA}/\sqrt{\mathrm{Hz}}$, while keeping a fast current modulation capability due to a control circuitry placed in parallel to the QCL and based on a field-effect transistor (FET).

The molecular transition chosen in this work is the P(29) of the $(0\;\;1^1\;\;1\;\;-0\;\;1^1\;\;0)$ rovibrational band of CO_2 at 2311.5152 cm^{-1}. The inset of Fig. 1 shows a typical scan of the PS signal at a pressure of 8.9 Pa, when the laser frequency is tuned across the molecular resonance. By carefully balancing the differential detection, a zero-offset signal is obtained. It ensures a linear conversion of the laser frequency fluctuations into amplitude variations in the region centered around the resonance frequency.

For the QCL frequency stabilization, the PS signal is processed by a home-made proportional–integral–derivative (PID) controller and fed back to the FET gate for current control. From a preliminary analysis of the free-running frequency-noise power spectral density (FNPSD) of a similar QCL [16], a kilohertz-level linewidth should require a locking bandwidth of about 100 kHz. In order to ensure this condition, we designed both the differential amplifier and the PID to have bandwidths larger than 1 MHz. However, there are two more fundamental aspects that can limit the loop bandwidth. The first is the roll-off of the QCL tuning rate with the modulation frequency, for which we confirm the results presented in [8]: the tuning rate is never flat, even at low frequencies, and shows a –3 dB cutoff at about 100 kHz. The second is the width of the linear region of the PS signal, which introduces a frequency roll-off starting from 300 kHz. Following the above considerations, the bandwidth of the frequency-locking loop is expected to be in the range of a few hundred kilohertz.

In order to characterize the frequency lock, two different measurements are carried out in parallel. The first one is the spectral analysis of the in-loop PS signal, the second one is the analysis of the beat note between the QCL and a narrow mid-IR coherent source [17] providing a stable (10 Hz linewidth within 100 μs) and absolute reference. Each measurement has also been performed with the QCL in the free-running regime.

In Fig. 2 the FNPSD measurement results are shown. First, it is noteworthy to highlight the improvements in the free-running regime brought by the evolution of the driver [traces (a) and (b)]: the FNPSD now exhibits a clean $1/f$ trend, confirming that virtually no external noise is added. By closing the frequency-locking loop, the FNPSD is reduced in the spectral range below 250 kHz [trace (d)], which is then assumed to be the loop bandwidth, as expected. At about 450 kHz, the onset of a self-oscillation peak is evident. It can be explained well by the dephasing introduced by the approaching roll-offs mentioned above, and it is, at present, the factor limiting the loop performances. The FNPSD of the locked QCL [trace (c)] is obtained by adding to the closed-loop error signal [trace (d)] the detection noise floor [trace (e)]. The latter is dominated, in the low-frequency range, by the residual intensity noise of the QCL and limits the frequency-noise reduction.

The effect of the locking on the QCL emission lineshape can be more intuitively described by the beat-note spectrum, as shown in Fig. 3. The 450 kHz servo bumps confirm the oscillation peak appearing in the FNPSD. By comparing the areas of the locked and free-running beat notes, we find that 77% of the QCL radiation power is forced into the narrow peak centered on the molecular line. Switching from the free-running to the locked regime, the linewidth (FWHM) is reduced from about 500 kHz down to 760 Hz on a 1 ms time scale (inset). The inset also shows the comparison between the beat note and the locked QCL power spectral profile retrieved from its FNPSD, following [18], over a 1 ms time scale (Fourier frequencies > 160 Hz). For the latter, a 900 Hz FWHM is obtained, in good agreement with the beat-note linewidth.

The beat-note frequency was also measured by a 1 s gated frequency counter over about 2 h. The obtained

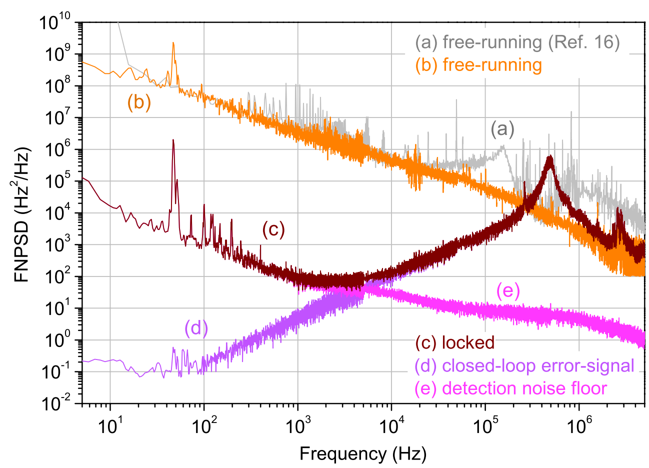

Fig. 2. (Color online) Traces (a) and (b): free-running QCL FNPSD with the former [16] and present versions of the home-made driver, respectively. Trace (c): locked QCL FNPSD, obtained by summing the spectrum of the closed-loop error signal [trace (d)] to the spectrum of the PS detection noise floor, measured with the empty gas cell [trace (e)].

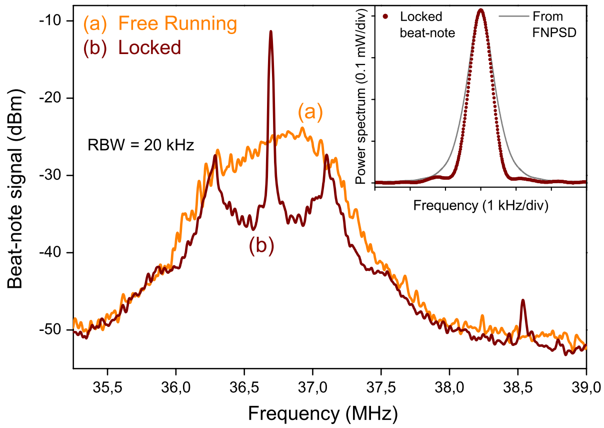

Fig. 3. (Color online) Fast Fourier transform spectra of a 10 ms long evolution of the beat note between the QCL, in free-running [trace (a)] and locked [trace (b)] conditions, and the narrow mid-IR source. Inset: zoomed view (linear scale) of the central peak observed over 1 ms with a resolution bandwidth of 721 Hz (dotted curve) and the QCL power spectral profile retrieved from the locked FNPSD (solid curve).

Allan deviation is 3 kHz at 1 s and decreases down to 0.9 kHz up to 320 s. Then, for longer times, it increases again, due to slow variations of the locking signal offset. This prevents our oscillator from achieving the stability performances of the best mid-IR standards [19].

The absolute frequency of the CO_2 line was measured by averaging a set of frequency counts performed over several days and in different experimental conditions. The obtained value is 69297478.776 ± 0.025 MHz, with an uncertainty that takes into account both the repeatability of the offset zeroing and the OFCS accuracy. This result is in agreement with the value given by the HITRAN database [20,21] for this transition, but with at least two orders of magnitude increased accuracy.

In conclusion, we demonstrate frequency locking of a mid-IR QCL to a secondary frequency standard (CO_2 line) by PS. A 760 Hz linewidth is obtained, and the QCL absolute frequency is stabilized to the center of the CO_2 line at a precision level of $4 \cdot 10^{-11}$ in 1 s. The absolute frequency of the locked QCL is measured with an accuracy of $4 \cdot 10^{-10}$. The locking bandwidth, at present limited by the modulation bandwidth of the QCL, can be increased by implementing alternative schemes, including external modulators or optical injection. On the other hand, phase locking of the QCL directly to an OFCS could improve the long-term stability.

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