Quantum cascade laser-based mid-IR frequency metrology system with ultra-narrow linewidth and 1×10^{-13}-level frequency instability

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We demonstrate a powerful tool for high-resolution mid-IR spectroscopy and frequency metrology with quantum cascade lasers (QCLs). We have implemented frequency stabilization of a QCL to an ultra-low expansion (ULE) reference cavity, via upconversion to the near-IR spectral range, at a level of 1×10^{-13}. The absolute frequency of the QCL is measured relative to a hydrogen maser, with instability $<1 \times 10^{-13}$ and inaccuracy 5×10^{-13}, using a frequency comb phase stabilized to an independent ultra-stable laser. The QCL linewidth is determined to be 60 Hz, dominated by fiber noise. Active suppression of fiber noise could result in sub-10 Hz linewidth.

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High-resolution spectroscopy in the mid-IR has been a crucial tool for the elucidation of fine details of molecular structure and dynamics for many decades. Earlier, mostly gas lasers (CO_2, CO, He–Ne) with narrow linewidth (kHz to sub-Hz, e.g., [1]) have been used as sources, permitting spectral resolutions below the Doppler width of gaseous room temperature samples, by employing appropriate nonlinear spectroscopic techniques. The molecular gas lasers are only line-tunable, in some cases severely restricting their applicability. As a remedy, the generation of tunable microwave sidebands has been implemented successfully [2], but the method has the drawback of the low power in the sidebands. Another approach is the difference-frequency generation (DFG) from near-IR sources which allows a wide tuning range, but generally suffers from low mid-IR power [3].

Quantum cascade lasers (QCLs) offer both high power and relatively wide tuning. The free-running linewidth on the timescale of 1 s is typically on the order of 1 MHz and has been the subject of several studies [4,5]. Linewidth narrowing, along with absolute frequency stabilization and, finally, absolute frequency measurement, are therefore important tasks for rendering QCLs usable for high-resolution spectroscopy.

It has already been shown that the QCL linewidth can be dramatically reduced by locking to a reference cavity [6], by phase-locking to a narrow-linewidth CO_2 reference laser, reaching less than 10 Hz relative linewidth [7], or by phase-locking to the DFG wave generated from two near-IR cw lasers [8]. This latter approach, however, limits the spectral coverage of the QCL, since it relies on particular reference lasers.

The upconversion approach [9] appears as particularly suitable as a general approach for QCL frequency metrology and linewidth narrowing [10]. References [8,10] also performed absolute frequency measurements. In our own work [11], we upconverted radiation from a 5.4 μm QCL to 1.2 μm by mixing with a cw 1.5 μm fiber laser. These two near-IR waves were simultaneously measured by and stabilized to a standard Er:fiber comb referenced to a hydrogen maser. No linewidth narrowing was implemented at the time.

In the present work, we extend significantly the performance of QCL frequency metrology by demonstrating absolute frequency stabilization, linewidth narrowing, and absolute frequency measurement at the 10 Hz level without relying on an ultra-stable mid-IR reference laser. An important advantage of the present approach is that it is applicable to any QCL in the range 5–12 μm.

In practice, a flexible solution for both linewidth narrowing and absolute frequency stabilization is provided by locking the QCL to an ultra-stable reference cavity made of ultra-low expansion glass (ULE). This approach is common for lasers emitting in the near-IR and visible spectral ranges. However, to the best of our knowledge, it has not yet been implemented with QCLs. The use of a reference resonator has the advantage that the QCL is stabilized without the need of a frequency comb, a significant simplification. For applications where the optical frequency of the QCL must also be measured or accurately monitored, a comb is nevertheless still required. For such cases, we propose and demonstrate here an approach where the upconverted radiation, rather than the QCL, is stabilized to a ULE reference cavity. This has the significant advantage that the reference cavity can use coatings for the near-IR and that less expensive and higher-performance near-IR components, rather than mid-IR optical components, can be employed.

The basic concept of the spectrometer for mid-IR frequency metrology has been described before [11]. In this Letter, the spectrometer has been further developed such that it provides fast frequency stabilization of a QCL and significant linewidth narrowing. The overall concept is as follows. A standard near-IR fiber-based frequency comb (pulse repetition frequency f_{rep}) is phase-locked to a ULE cavity (ULE1) stabilized laser. This provides a comb of ultra-narrow modes with Hz-level linewidths. A near-IR laser (frequency $f_{1.5}$) is phase-locked to the nearest mode of the frequency comb. The sum frequency (upconverted) wave of the near-IR laser and of the QCL (frequency $f_{1.2} = f_{1.5} + f_{\mathrm{QCL}}$) is frequency stabilized to another stable cavity (ULE2) by controlling f_{QCL}. Therefore, the QCL frequency f_{QCL} is also stabilized. Furthermore, the frequencies $f_{1.5}$, $f_{1.2}$, and f_{rep} are simultaneously measured by the comb and a hydrogen maser (H-maser) in real time to allow measuring f_{QCL} and its spectral properties, i.e., its fluctuations.

A schematic of the apparatus in shown in Fig. 1. An erbium-doped fiber-based frequency comb is phase-locked to a 1064 nm Nd:YAG laser which is stabilized to a compact and robust reference cavity (ULE1), achieving 1×10^{-15} short-term frequency instability [12]. The phase-lock acts on the cavity length of the frequency comb via an intracavity electrooptic phase modulator and a piezo-actuated cavity mirror, controlling the repetition rate such that the beat (Δ_{ULE1}) between the Nd:YAG laser (frequency f_{ULE1}) and a nearby comb mode (integer mode number m_{ULE1}) has a fixed value, Δ_{ULE1}. The residual linewidth of this beat is less than 1 Hz. The comb repetition rate f_{rep} is then determined by the frequency of the optical reference ULE1 through the phase-lock condition $m_{\mathrm{ULE1}} f_{\mathrm{rep}} \pm f_{\mathrm{CEO}} \pm \Delta_{\mathrm{ULE1}} = f_{\mathrm{ULE1}}$. The carrier envelope offset frequency f_{CEO} is stabilized to a fixed-frequency RF signal (20 MHz). In this way, all modes of the comb are of high absolute stability and ultra-narrow. Using a separate ultra-narrow 1.5 μm laser, we determined an upper limit of 6.5 Hz (resolution bandwidth limited) for the linewidth of comb modes at this wavelength.

The repetition rate f_{rep} is measured by a frequency counter referenced to the H-maser, with 1 s integration time. The frequency instability of the maser is below 2×10^{-14} for integration times longer than 10 s; its frequency is determined to an uncertainty 5×10^{-13} by comparison with GPS signals. The values of f_{rep} and f_{CEO}, combined with the frequency $\Delta > 0$ of the beat of a given laser wave (frequency f) and a comb mode (mode number m), yield the actual optical frequency, $f = m f_{\mathrm{rep}} \pm f_{\mathrm{CEO}} \pm \Delta$, where m and the sign associated with Δ are determined using a wavemeter having suitable accuracy. The sign associated with f_{CEO} is determined by the corresponding phase-lock circuit.

The employed QCL is a cw room temperature, distributed feedback grating, ridge waveguide QCL (Maxion). A two-stage temperature stabilization system allows the QCL to be operated between −10°C and +13°C, corresponding to a tuning range from $\lambda_{\mathrm{QCL}} = 5350$ to 5368 nm, with an output power of 40 mW. For sum frequency generation, its radiation is overlapped with radiation from an Erbium-doped fiber-amplifier (EDFA), which is seeded by a 1.5 μm fiber laser and then sent to an orientation-patterned (OP) GaAs crystal. 30 μW of the sum frequency wave at 1.2 μm are delivered via a 2 m long fiber to an actively vibration isolated breadboard carrying a second ULE cavity (ULE2). An AOM (AOM1) is used to suppress interferences between the fiber and the ULE cavity, and a circulator guides the wave reflected from the cavity onto a photodiode for error signal detection. The ULE cavity spacer is 8.4 cm long and the cavity finesse is 160 000. A two-stage temperature stabilization system acts on the cavity block inside the vacuum chamber and the chamber itself. The 1.2 μm radiation is stabilized to the cavity by the Pound–Drever–Hall technique by controlling the frequency of the QCL. The 1.2 μm radiation is frequency modulated by modulating the QCL current at 9.6 MHz through a bias-tee, which is also used for the fast frequency feedback signal. The slow frequency feedback signal is applied to the modulation input of the current driver.

The zero-order output of AOM1 is led through an optical isolator and then coupled into an optical amplifier which boosts the power level of the 1.2 μm radiation to approx. 0.1 mW. This wave is then sent via a 30 m long fiber to the frequency comb, where it produces a beat Δ_2 with a comb mode.

The 1.5 μm fiber laser is phase-locked to the frequency comb such that the beat frequency Δ_1 of the fiber laser and the comb is constant by means of an AOM (AOM2) and the piezo inside the laser.

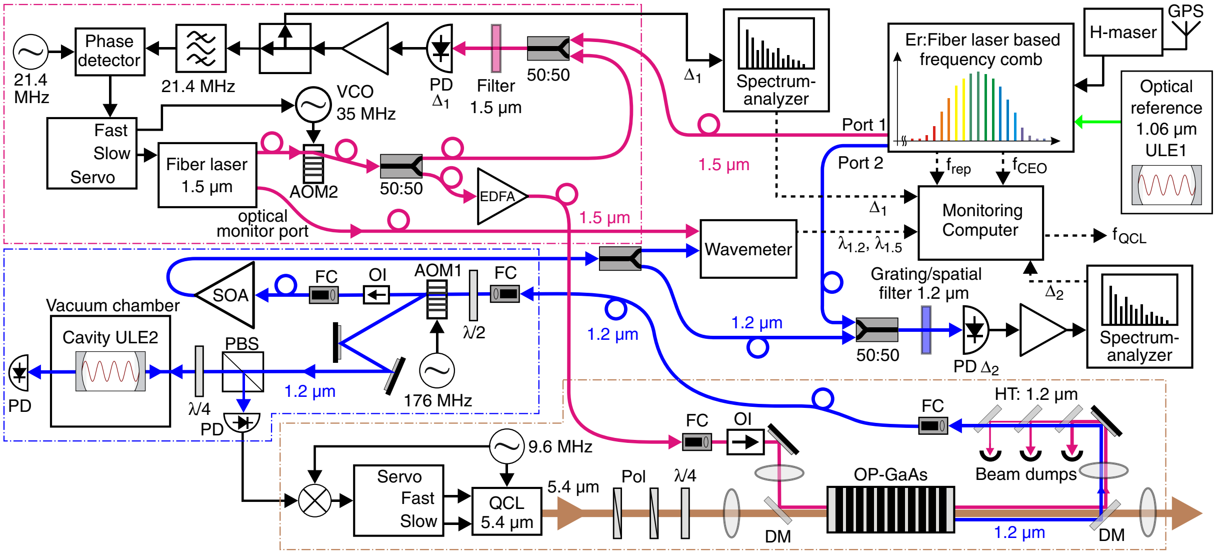

Fig. 1. Schematic of the setup. Pink: fiber laser wave at 1.5 μm; brown, QCL wave at 5.4 μm; blue, sum frequency wave at 1.2 μm. Full lines, analog electric signals; dashed lines, digital signals. DM, dichroic mirror; AOM, acousto-optic modulator; SOA, semiconductor optical amplifier; Pol, wire grid polarizer; PD, photodetector; OI, optical Faraday isolator; FC, fiber collimator; PBS, polarizing beam splitter; VCO, voltage-controlled oscillator. The spectrum analyzers are referenced to the H-maser.

Then, $f_{1.5} = m_1 f_{\text{rep}} \pm f_{\text{CEO}} \pm \Delta_1$. The fiber laser is connected to the frequency comb using a second 30 m long fiber. A spectrum of the beat Δ_2 between the frequency comb and the 1.2 μm wave is shown in Fig. 2, the FWHM is 11 Hz. The beat Δ_1 of the fiber laser with the comb line to which it is phase-locked is also shown in the figure; it has sub-Hz linewidth. The two measured linewidths need to be corrected for the influence of the fiber noise of the two 30 m long fibers, which are not actively stabilized. The fiber noise was measured by sending the 1.5 μm laser wave in a round-trip through the two fibers and performing a self-beat. This resulted in 120 Hz FWHM. This implies that at the location of the 1.5 μm laser, its linewidth is on the order of 60 Hz. On the other hand, the linewidth of the 1.2 μm wave is expected to be substantially less than 11 Hz, since the 1.2 μm light used for making a beat with the comb has been broadened by propagation through the 30 m long fiber. The linewidth of the QCL is not observed directly, but can be inferred from the geometric sum of the 1.2 μm and the 1.5 μm linewidths, to be approximately 60 Hz, dominated by the fiber noise on the 1.5 μm wave. Active suppression of the fiber noise could result in linewidth narrowing of the QCL to the sub-10 Hz level, i.e., to the inferred linewidth of the 1.2 μm wave at the ULE2 cavity.

The absolute frequency properties of the QCL are determined as follows. The two near-IR frequencies are given by $f_{1.2} = m_2 f_{\text{rep}} \pm f_{\text{CEO}} \pm \Delta_2$ and $f_{1.5} = m_1 f_{\text{rep}} \pm f_{\text{CEO}} \pm \Delta_1$, where m_1 and m_2 are the respective constant mode numbers computed from wavemeter readings, and the values of Δ_1 and Δ_2 are measured by spectrum analyzers (2 s integration time) every 5 s via peak detection. The QCL's optical frequency is then computed as $f_{\text{QCL}} = (m_2 - m_1) f_{\text{rep}} \pm (\Delta_2 \mp \Delta_1)$ by the measurement computer in real time.

Note that both the upconverted waves frequency and the QCLs frequency are essentially determined by the frequencies of the ULE cavities. While $f_{1.2}$ is directly given by a cavity mode frequency of ULE2 ($f_{1.2} = f_{\text{ULE2}}$), f_{QCL} is determined by cavity mode frequencies of both ULE1 and ULE2, according to $f_{\text{QCL}} = -(m_1/m_{\text{ULE1}}) \times f_{\text{ULE1}} + f_{\text{ULE2}} \quad -(m_1/m_{\text{ULE1}})(f_{\text{CEO}} \mp \Delta_{\text{ULE1}}) \quad -f_{\text{CEO}} \mp \Delta_1$. We recall that f_{CEO}, Δ_{ULE1}, Δ_1 are constant RF frequencies. Therefore, the frequencies $f_{1.2}$ and f_{QCL} will be affected by the (small) drifts of the ULE cavities' frequencies due to temperature variations by mechanical relaxation, as well as by locking errors.

A measurement of the scaled comb repetition rate $(m_2 - m_1) f_{\text{rep}}$ as a function of time is shown in Fig. 3. Its drift is due to the counter noise of ULE1, which we observed to vary slowly with time. The frequency instability, due to counter noise and H-maser frequency instability, is 5 Hz on the 1 s time scale, and averages down to ≤1 Hz at integration times >10 s (after drift removal).

The upconverted waves frequency and QCLs frequency are also presented in Fig. 3. The frequencies $f_{1,2}$, f_{QCL} have small drifts of 0.05 Hz/s and 0.01 Hz/s, respectively. These drifts are not constant in time, but are typically at this level and may be attributed to the reference cavities ULE1 and ULE2. We emphasize that the QCL frequency is measured in real time and, therefore, in an actual spectroscopic experiment it will be possible to correct for the observed drift either by active control, or during the data analysis. The frequency instability of the QCL is 5 Hz (1×10^{-13} in fractional terms) for averaging times larger than 10 s (after drift removal). The small systematic deviations of the QCL frequency are of order 30 Hz; the above determination of the instability of $(m_2 - m_1) f_{\text{rep}}$ and the negligible instability of the beat Δ_1 indicates that the deviations are not dominantly due to the comb and fiber laser but caused by residual errors of the QCL lock to cavity ULE2, i.e., they are expressed via the Δ_2 contribution to f_{QCL}. The absolute optical frequencies at $t = 0$ s were measured to be $f_{1.2,0} = 247.4776695293(2)$ THz and $f_{\text{QCL},0} = 55.91676292253(3)$ THz. The uncertainties are due to the H-maser frequency uncertainty.

In conclusion, we have demonstrated both absolute frequency stabilization at a level of 1×10^{-13} (after linear drift removal) and linewidth narrowing of a standard QCL to the 60 Hz level (1×10^{-12}). Absolute frequency

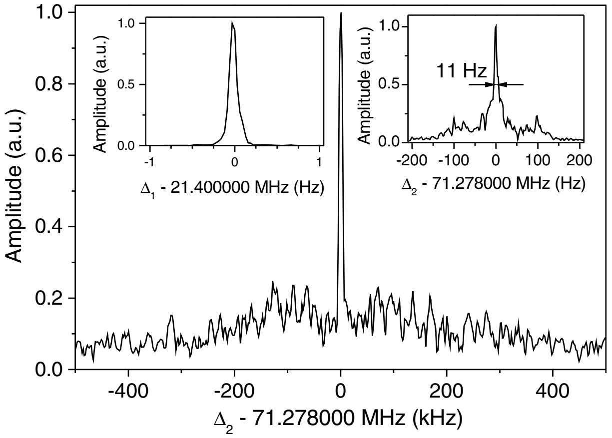

Fig. 2. Beat note Δ_2 of the 1.2 μm upconverted wave with a mode of the frequency comb. The wave is not locked to this mode. Resolution bandwidth (RBW) 6.4 kHz, sweep time 21 ms. Left inset: beat note Δ_1 of the 1.5 μm laser with a mode of the frequency comb. The laser frequency is phase-locked to this mode, FWHM 0.1 Hz, RBW 31.25 mHz, sweep time 32 s. Right inset: high-resolution measurement of the 1.2 μm beat, 11 Hz FWHM, RBW 6.4 Hz, sweep time 2.2 s.

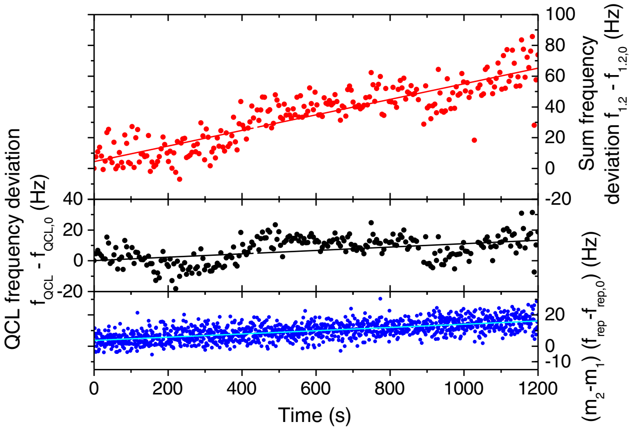

Fig. 3. Simultaneous frequency measurement of the scaled comb repetition rate $(m_2 - m_1) f_{\text{rep}}$, of the upconverted wave's $f_{1.2}$, and of the QCL's f_{QCL}, respectively. Lines: Linear fits. $(m_1 - m_2) f_{\text{rep},0} = 55.916742336214(5)$ THz.

measurement is also achieved, with uncertainty of 30 Hz (5×10^{-13}). The two key ingredients are a high-finesse ULE cavity for the near-IR and an optically referenced near-IR frequency comb. The present system appears well suited for ultra-high resolution spectroscopy, e.g., of cold, trapped molecules. The QCL linewidth is broadened by fiber noise, but this may be eliminated in a straightforward manner in future work and would result in sub-10 Hz linewidth.

The described approach has the advantage that it combines strong linewidth narrowing and absolute frequency stabilization with absolute frequency measurement. The approach is general, since the employed nonlinear crystal (GaAs) is suitable for upconversion of QCL wavelengths covering the whole mid-IR. However, alternative crystals could also be used. We note that a first convenient simplification would consist in stabilizing the 1.5 μm laser to a ULE cavity and using the laser both for upconversion of the QCL and as a reference laser for stabilization of the comb. A second simplification is to use only a single near-IR ULE cavity, with appropriately designed mirror coatings, instead of two cavities.

One additional outcome of this work is the capability of observing the linewidth of the QCL in real time via the observation of the beat between the upconverted wave and a comb mode, provided that fiber noise in the connection between the fiber laser, the generated sum frequency wave and the frequency comb is either actively suppressed or small compared to the observed linewidth. The other sources of linewidth broadening, upconversion and comb, contribute negligibly to the beat linewidth.

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