Mid-infrared multiheterodyne spectroscopy with phase-locked quantum cascade lasers

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Fabry-Pérot (FP) quantum cascade lasers (QCLs) provide purely electronically controlled monolithic sources for broadband mid-infrared multiheterodyne spectroscopy (MHS), which benefits from the large gain bandwidth of the QCLs without sacrificing the narrowband properties commonly associated with the single mode distributed feedback variant. We demonstrate a FP-QCL based multiheterodyne spectrometer with a short-term noise-equivalent absorption of $3 \times 10^{-4} / \sqrt{\text{Hz}}$, a mid-IR spectral coverage of 25 cm⁻¹, and very short acquisition time (10 μs) capability. The broadband potential is demonstrated by measuring absorption spectra of ammonia and isobutane under atmospheric pressure conditions. The stability of the system is enhanced by a two-stage active frequency inter-locking procedure, where the two QCLs are pre-locked with a slow feedback loop based on an analog frequency discriminator, followed by a high bandwidth optical phase-locked loop (OPLL). The locking system provides a relative frequency stability in the sub kHz range over seconds of integration time. The strength of the technique lies in the ability to acquire spectral information from all optical modes simultaneously and individually, which bodes for a versatile and cost effective spectrometer for mid-IR chemical gas sensing.

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The mid-infrared part of the electromagnetic spectrum contains many fundamental rotational-vibrational transitions of molecules that have important roles in environmental studies, emission monitoring, medical diagnostics and security applications\(^1\)\(^-\)\(^5\). This spectral region has therefore received much attention over the years and several spectroscopic absorption techniques have been proposed and developed, each with their strengths and weaknesses. Fourier transform infrared spectroscopy (FTIR) is perhaps the most widely adopted and has been applied in numerous measurement applications during the last decades. Although it is unrivaled in terms of spectral coverage, it utilizes opto-mechanical moving parts, which makes it inherently slow, with acquisition times on the order of seconds to minutes depending on spectral bandwidth and resolution. Therefore, other techniques have emerged for in-situ broadband mid-IR gas concentration assessments, where the response time of the system needs to be in the single second regime or even faster. Such measurements may involve transient chemical reactions or real-time process control, where slow acquisition either struggles to resolve the transient phenomena or provides a feedback that is too slow for active control of the process.

Laser spectroscopy provides means for performing spectroscopy with high temporal resolution, but historically the light sources have been restricted by their tuning capabilities and narrow spectral coverage. Recent developments of external cavity (EC) systems\(^6\) and devices with integrated heaters\(^7\)\(^,\)\(^8\) aim to fully utilize the gain bandwidth of the lasers, but they rely on intricate device design or moving parts to cover the entire optical bandwidth. A promising alternative is provided by the development of optical frequency comb sources, primarily in the near-infrared (NIR) spectral region, and frequency comb-based spectrometers are nowadays frequently used in various spectroscopic applications\(^9\)\(^-\)\(^17\).

It was recently demonstrated that also multimode Fabry-Pérot quantum cascade lasers (FP-QCL) exhibit comb-like operation under certain conditions\(^18\), which makes QCL-based multiheterodyne spectroscopy measurements in the dual-comb spectroscopy\(^19\) (DCS) configuration feasible. There are many potential benefits of a mid-IR spectrometer based on this approach: the aforementioned fast response time, or the potential for integration of the light sources on a single chip\(^20\). Especially the latter property could reduce both cost and physical dimensions of spectroscopic sensor systems. These light sources are fully electronically controlled, with no moving parts, which potentially increases the stability of the system and reduces the maintenance demand. It is however important to note that a chemical gas sensor of this type is unlikely to match an FTIR or a stabilized NIR dual-comb spectrometer in terms of spectral coverage, but it may be able to provide a favorable performance-to-price ratio for certain sensing applications that require moderately broadband, high resolution spectroscopic measurements in the mid-IR molecular fingerprint region.

The multiheterodyne spectrometer used in this work (schematically depicted in Fig. 1) is based on the DCS scheme\(^21\), incorporating two multimode optical FP-QCL sources with slightly different FP mode spacings (spacing difference of
A 160 MHz was used in this experiment). Through multiheterodyne mixing of the two sources, the optical absorption and dispersion information is conveniently down-converted to the radio frequency domain for measurement. The two FP-QCLs are a part of a single chip\(^{22,23}\) (separated by 500 \(\mu\)m) and share a common Peltier element for thermal tuning. Two laser drivers (Wavelength Electronics QCL500 and QCL1000) were used to supply injection currents to both lasers separately. The laser chip is housed in a water-cooled custom laser mount with a single large diameter (1 inch) collimation lens (f/0.6). After collimation, the beams are directed through a set of two irises (not shown in the diagram) to ensure that any reflections from the succeeding optical surfaces in the system do not provide unwanted feedback that could perturb the operation of the lasers. Mitigation of the optical feedback has proven crucial for stable multiheterodyne beat notes during data acquisition.

The beams from the two FP-QCLs are split into two arms by ZnSe plate beamsplitters. This yields four mid-IR beams (two from each laser), and two additional beam splitters are used to overlap the two beams (one from each laser) onto two fast photodetectors that are labeled as sample and reference detectors. In the sample detector channel, one laser (QCL1, referred to as signal laser) interacts with the sample gas (12 cm absorption cell) and is then incident on the detector (see left part of Fig. 1). In the reference detector channel, the signal laser is directed to the detector without interaction with the sample. Another laser (QCL2 in Fig. 1) acts as a local oscillator for both detector channels. Since the reference detector signal is unaffected by the sample gas, it is used for optical power normalization\(^{21}\).

The beams are focused by off-axis parabolic mirrors onto two thermoelectrically cooled photodetectors from VIGO System S.A. (PVI-4TE-10.6 with no immersion lens, field of view FOV = 70\(^{\circ}\), and tilted detector windows to reduce optical feedback). The detectors have an active area of \(0.03 \times 0.03\) mm\(^2\), a \(D^*\) of \(2 \times 10^8\) cm-Hz\(^{1/2}\)/W and a 3 dB cutoff frequency of 1.1 GHz, which enables detection of up to 18 multiheterodyne beat notes with our laser pair using the beat note folding procedure\(^{21}\). A FP mode spacing of 1.36 cm\(^{-1}\) results in an instantaneous spectral coverage of ~25 cm\(^{-1}\). The RF beat notes are amplified by low-noise RF amplifiers (HD Communications HD28110) and measured by two real time spectrum analyzers from Tektronix (RSA5103A /RSA6106A). An acquisition rate of 25 MS/s with a bandwidth of 25 MHz around the center beat note frequency was used to determine the relative absorption for each beat note compared to the reference.

The measurement procedure used to produce the absorption spectra was the following: (1) a gas mixture containing the analyte was introduced to the sample cell, (2) data acquisition from the signal detector was performed by stepping through the center frequencies of the beat notes, (3) the same procedure was performed for the reference detector. After complete data acquisition the signals were analyzed in post-processing by comparing the amplitude versus time traces in the sample and reference channels. A Rician distribution\(^{24,25}\) of the beat note amplitudes was assumed in the data analysis algorithm to calculate the transmission spectrum. Due to the bandwidth and buffer-size limitations of our spectrum analyzers the beat
Notes were measured sequentially by recording a 10 μs time trace for each beat note. This was sufficient to study the system in terms of short-term noise performance. These limitations are purely technical and will be addressed in the future by implementing a fast digitizer with an acquisition bandwidth matched to the detector bandwidth combined with high bit-rate data streaming and real-time computing, which should provide simultaneous IQ-demodulation of all beat notes and enable long-term data acquisition.

To perform reliable spectroscopic measurements, it is important to prevent drifts of the multiheterodyne beat note frequencies, which would significantly complicate the signal processing and potentially introduce errors to the measurements. A pair of free-running FP-QCLs can exhibit intermutual frequency drifts in the MHz range over minute time scales, which can be reduced by actively stabilizing the two FP-QCLs relative to each other (inter-locking) using a frequency discriminator circuit. This ensures that the multiheterodyne RF beat notes remain at fixed frequency positions for the duration of the data acquisition. If necessary, an additional absolute frequency lock can be realized by conventional techniques using an external frequency reference, such as a well-defined absorption transition, a frequency stabilized coherent source, or a reference high-finesse cavity.

To further improve the mutual frequency stabilization and to reduce the effective linewidth of the heterodyne beat notes, a two-stage hybrid inter-locking system shown schematically in Fig. 1 has been developed. The RF signal from the reference photodetector, containing the beat signals from the optical interaction between the two laser beams, is split into two parts, feeding (1) a slow frequency discriminator-based feedback loop as described below, and (2) a fast optical phase locked loop (OPLL) circuitry, respectively.

FIG. 1. Schematic of the hybrid inter-locking system. The RF signal from the transimpedance amplifier of the photodetector is divided into two parts: (blue) accounts for slow frequency drifts with a large frequency locking range (locking bandwidth <100 kHz), (red) corrects fast phase fluctuations by an OPLL with a bandwidth limited by the analog modulation input of the laser current driver (<3 MHz).

The signal in the analog frequency discriminator part is first amplified by a low noise (NF<0.8 dB) 40 dB RF amplifier (Pasternack Inc. PE15A1012) followed by band-pass filtering around 40 MHz, which corresponds to a quarter of the FSR.
difference (the frequency of the first beat note available in the RF spectrum). The frequency to amplitude transduction is obtained by a custom-made Pi topology 9th order Butterworth LC low pass filter (LPF) with a cutoff frequency slightly lower than 40 MHz, which provides a nearly linear sharp attenuation slope (2.3 dB/MHz) around the locking frequency [see Fig. 2(a)]. An RF gain detector (Analog Devices AD8302) compares the signal levels of the stabilized RF beat note after passing through the low-pass filter (frequency-dependent attenuation), with that of a flat frequency response attenuation of 20 dB (Mini-Circuits HAT-20+). Within the RF gain detector chip, the measured difference between the two signals after logarithmic amplification yields an intensity-independent output that provides an error-signal proportional to the frequency deviation of the beat note of approximately 70 mV/MHz in a linear scale. An analog PID controller (SRS SIM960, 100 kHz bandwidth) is supplied with the error-signal to actively control the injection current of one of the signal laser (QCL1 in Fig. 1) through the modulation input of the laser driver.

The bandwidth of the PID controller (<100 kHz) is only sufficient to provide a coarse beat note frequency stabilization, but is not sufficient to fully inter-lock the two lasers and narrow the beat note linewidth. For this purpose, an optical phase locked loop circuit (OPLL) is used in parallel to perform inter-locking with higher bandwidth. This was realized by incorporating a fast OPLL system commercially available from Toptica, Inc. (mFALC 110). The mFALC 110 comprises a fast analog PID controller and an RF phase detector, which mixes the RF signal from the photodetector with a stable RF reference from an external source (SRS SG384). This process results in a frequency dependent error signal, which is used to actively control the injection current of the reference laser (QCL2 in Fig. 2). The laser current driver bandwidth of ~3 MHz determines the effective control loop bandwidth. A clear indication of phase-locking is observed with the OPLL [see Figure 2(b)], which considerably improves the signal-to-noise ratio of the multiheterodyne beat notes.

The use of the OPLL by itself proved to be unstable due to its narrow capturing range, which required the beat note to be in close proximity to the desired frequency to enable a successful lock. This issue was circumvented by the application of the slower analog frequency discriminator loop along with the OPLL. This hybrid inter-locking scheme provided reliable phase-lock and stabilization of the two FP-QCLs over extended timescales (hours).
FIG. 2. (a) The frequency response of the 9th order Butterworth LC low pass filter used in the frequency discriminator locking system. (b) RF beat note measured at 40 MHz (RBW = 100 Hz) with and without hybrid inter-locking enabled. The addition of the OPLL concentrates the beat note power to a narrow bandwidth, which considerably increases the signal-to-noise ratio.

The spectral distribution of the phase-locked beat note signal shows that the signal energy is concentrated around the peak [see Fig. 2(b)], which is similar to that reported in earlier works on single-mode QCLs and Josephson oscillators. 3 dB widths of the beat notes (FWHM) in the range of ~11 – 220 kHz was observed, where the values in the lower part of the range were observed for beat notes in proximity to the locking beat note. This provides a signal-to-noise of >40 dB, which is a significant improvement over the analog frequency discriminator locking circuit alone that resulted in a Gaussian-shaped beat note with a signal-to-noise ratio of ~23 dB and a FWHM of 2.3 MHz. It should be noted that while the linewidths of the beat notes are narrowed, only the relative frequency of the two lasers is stabilized, and the linewidths of the corresponding optical modes are not reduced.

FIG. 3. (a) A time series measurement of the center frequency of the RF beat note at ~40 MHz for the different beat note stabilization scenarios. The center frequency was evaluated by fitting a Gaussian profile to the beat note and extracting the center frequency from the fit. (b) Allan deviation plot using the data from (a).

The locking performance of the multiheterodyne system was characterized by measuring the center frequency of the beat note at ~40 MHz as a function of time [see Fig. 3(a)]. The corresponding Allan deviation plot is shown in Fig. 3(b). As can be seen in the figure, the free-running system suffers from a large drift of the RF beat notes’ center frequency, visible in the blue trace of Fig. 3(a). The introduction of the analog frequency discriminator locking system increased the stability, showing a 1σ Allan deviation of 12 kHz for 10 s of integration time. Finally, the hybrid inter-locking scheme improved the frequency stability further by more than an order of magnitude down to ~400 Hz observed at 10 s integration time. The high frequency stability significantly simplifies the signal processing procedure by reducing the frequency jitter and drift of the RF beat notes, which will be a requirement for long term averaging as indicated in Ref. 28.
FIG. 4. (a) MHS measurements of ~50% ammonia in nitrogen mixture at atmospheric pressure (red markers) together with a calculated spectrum based on the HITRAN database\textsuperscript{29} (blue line) for the measurement conditions used. The number of available beat notes in this measurement gives a spectral coverage of ~25 cm\textsuperscript{-1}, mainly limited by a combination of the bandwidth of the photodetectors and the relatively large mode spacing difference of the FP-QCLs. The lower panel shows the residuals with a standard deviation of 0.105. (b) ~15% Isobutane measurements at atmospheric pressure (red markers) together with a calculated spectrum based on the PNNL database\textsuperscript{30} (blue line) for the measurement conditions used. The lower panel shows the residuals with a standard deviation of 0.016. (c) Beat note amplitude as a function of time for short time scales, acquired using the different frequency inter-locking schemes described in this work. The traces have been vertically offset for visualization purposes. (d) Allan deviation of the relative beat note amplitudes based on the traces in (c).

Spectroscopy has been performed using 18 optical modes detectable within the photodetector bandwidth (two modes at the highest frequencies were not analyzed; although clearly visible, they were too far outside the detector high frequency cut-off, which would require longer averaging to provide SNR consistent with other modes). Figure 4(a) shows a measurement of ~50% ammonia (NH\textsubscript{3}) in nitrogen mixture at atmospheric pressure (red circles). Ammonia is a small molecule and its spectrum exhibits multiple narrow spectral features within the tuning range of the laser as showed by a spectrum simulated for the actual experimental conditions using the HITRAN database\textsuperscript{29} [blue line in Fig. 4(a)]. The data was recorded with an acquisition time of 10 μs per mode and shows good agreement with the simulated spectrum. The error bars in Fig. 4 indicate the 1σ confidence interval for the measured values calculated based on the error propagation (see details in Supplementary material). The bandwidth normalized noise-equivalent absorption\textsuperscript{13} (NEA) was determined as the standard deviation of the fit residual (see Supplementary material), which gives an NEA of ~3×10\textsuperscript{-7}/√Hz for the ammonia measurement. It should be noted that the primary source of the clearly noticeable discrepancies with the models are likely related to optical fringes in the system that interfere with the optical power normalization procedure. This translates into the fit-residual errors used for the
NEA calculation, and results in an upper bound estimate of the system sensitivity. Despite that, the hybrid inter-locking allowed the NEA to be improved by a factor of ~5 with respect to our earlier system not equipped with the OPLL. The current system also provides the capability of broadband spectral feature detection with NEAs comparable to earlier multiheterodyne systems using fast frequency scanning in conjunction with wavelength modulation spectroscopy or dispersion spectroscopy techniques.

Since the MHS system presented here retrieves the spectroscopic information primarily through a measurement of the beat note amplitudes, an analysis of the beat note amplitude stability has been performed to investigate the influence of the different frequency locking procedures and establish the ultimate short-term noise performance of the system. Figure 4 (c) shows the beat note amplitude as a function of time for the system operated in free-running mode (blue trace), slow feedback mode (red trace) and hybrid inter-locking mode engaging the OPLL (green trace). The corresponding Allan deviation plots are shown in in Fig. 4(d). A clear improvement in amplitude stability is observed for the frequency-stabilized systems with respect to the free-running lasers. The increased relative stability provided by the OPLL is mainly due to its higher bandwidth that concentrates the beat note power into a narrower bandwidth, which increases the carrier-to-noise ratio and allows to narrow the detection bandwidth without loss of spectroscopic information. Within the random noise limited regime indicated with a dotted line in Fig. 4(d) one can estimate an ultimate NEA of $2.8 \times 10^{-5}/\sqrt{\text{Hz}}$, which is highly encouraging in terms of the potential of the technology and indicates that the main limitation of the current system (NEA of $3 \times 10^{-4}/\sqrt{\text{Hz}}$) is not dictated by the noise in the multiheterodyne conversion process.

To also address measurements of large molecules with broadband rotationally unresolved spectra, an example measurement of isobutane ($\text{C}_4\text{H}_{10}$) was performed. A spectrum of ~15% isobutane ($\text{C}_4\text{H}_{10}$) in nitrogen mixture under atmospheric pressure conditions (red circles) together with a spectrum simulation based on the PNNL database (blue line) are shown in Fig. 4 (b). The shape of the broadband absorption feature of isobutane can clearly be resolved in a single spectral snapshot acquired during 10 μs by the FP-QCL based multiheterodyne spectrometer. A NEA of $5 \times 10^{-5}/\sqrt{\text{Hz}}$, calculated for this measurement, is close to the ultimate NEA estimated from Fig. 4(d), which indicates significantly smaller issue with optical fringe drift during the data collection compared to the ammonia measurement in Fig. 4(a).

In conclusion, a broadband mid-IR multiheterodyne spectrometer based on phase-locked FP-QCLs has been demonstrated by measuring absorption spectra from ammonia ($\text{NH}_3$) and isobutane ($\text{C}_4\text{H}_{10}$) at atmospheric pressure with a temporal resolution of 10 μs and short-term NEA ranging between $3 \times 10^{-4}/\sqrt{\text{Hz}}$ and $5 \times 10^{-5}/\sqrt{\text{Hz}}$. The two FP-QCLs are intermutually frequency stabilized by a hybrid inter-locking procedure, which uses a frequency discriminator for pre-locking.
and an OPLL for high bandwidth phase-locking yielding beat note linewidths of <220 kHz. The mid-IR multiheterodyne spectrometer can resolve up to ~20 RF beat notes (limited by the detector cut-off of <1.1 GHz), resulting in an instantaneous frequency coverage of ~25 cm⁻¹ around 8.5 μm. A reduced mode spacing difference would result in more densely packed RF beat notes and increase the spectral coverage accordingly. The combination of broadband coverage and fast acquisition time indicates that the system might be useful when assessing concentrations in transient chemical reactions, such as combustion processes. It has been shown previously that a FP-QCL based spectrometer can operate in single beat note detection mode, employing the same noise suppression modulation techniques that are commonly used for single mode lasers. This type of fully electronically controlled spectrometer can thus seamlessly switch between scanned narrowband detection, targeting narrow absorption features, or broadband detection covering the entire emission spectrum of the lasers. This can be accomplished with relatively low-cost FP-QCLs integrated on a single chip, which enables a versatile spectroscopic tool for mid-IR chemical gas sensing.

Supplementary Material

See supplementary material for the derivation of the 1σ confidence intervals (error bars) for transmission measurements in dual-detector multiheterodyne spectroscopy.

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