Terahertz multiheterodyne spectroscopy with quantum cascade lasers – a feasibility study

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***Abstract*—We study the feasibility of dispersion compensated terahertz quantum cascade lasers combs operating around 2.65 – 2.95 THz to perform multiheterodyne spectroscopy. The devices show short-term intermode beat note frequency drift after 30 μs, which indicates that reliable multiheterodyne spectroscopy over extended time-scale requires phase and timing correction to allow for coherent averaging.**

I. INTRODUCTION

MUTLIHETERODYNE spectroscopy with terahertz quantum cascade lasers (QCLs) arranged in the dual comb configuration has recently attracted attention due to its potential for molecular species identification in the terahertz spectral domain [1]. Much of this work is based on pioneering work performed in the mid-infrared [2], [3], where the maturity of the QCL-sources allows for room temperature operation with optical powers exceeding hundreds of milliwatts. Multiheterodyne spectroscopy (MHS) in the terahertz (THz), however, still possesses numerous challenges. In particular, the THz QCLs require cryogenic temperatures to operate, even with relatively modest optical powers in the sub-milliwatt range. Cryogenic temperatures are also required for the sensitive superconductive THz detectors.

In this work, we study the feasibility of THz-MHS for the purpose of spectroscopic assessments of gaseous compounds primarily related to security and safety applications.

II. EXPERIMENTAL PROCEDURE

Figure 1 shows the experimental setup, where two THz QCL with a slight mismatch of the cavity lengths (see Ref. [1]) are cooled to 37 K and cw-biased to operate in the comb regime. A closed cryo-system from ColdEdge Technologies based on a vibrationally dampened Gifford-McMahon (GM) cryocooler is used to provide large cooling capacity with relatively small mechanical vibrations. Low-noise laser drivers (Wavelengths Electronics, QCL2000+) are utilized to minimize the uncorrelated frequency and amplitude noise of the THz modes.

The laser emissions generated from the lasers are collimated by silicon lenses and off-axis parabolic mirrors. A HRFZ-Si beamsplitter combines the two beams that are overlapped on a liquid helium-cooled hot electron bolometer (HEB, Scontel), which provides the optical heterodyne mixing. This results in a multiheterodyne RF spectrum with multiple beat notes, regularly spaced with a spacing defined by the cavity length mismatch. MHS effectively down-converts the optical information from the two lasers to the RF domain in a parallel fashion [4], which enables further digitization and processing.

The comb-nature of the light sources can be evaluated by characterizing the intermode beating that occurs at a frequency corresponding to the free-spectral range (FSR) of the laser cavity [3]. The intermode signal can be extracted through optical mixing of the THz modes measured by the HEB or directly from the laser voltage through high-frequency bias-tees [5].

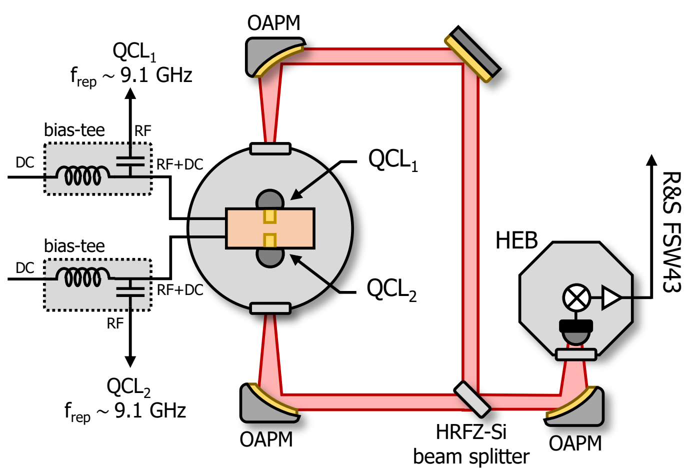

Fig. 1. Experimental setup. A vibrationally dampened cryostat houses the two THz QCLs, which are mounted in an anti-parallel configuration. The QCLs are biased through 12.5 GHz (1.5 A) bias-tees, whose RF outputs are measured by a real-time spectrum analyzer (R&S FSW43). The THz radiation is combined on a HRFZ-Si beamsplitter and focused on an HEB.

The multiheterodyne signals were acquired using the HEB in combination with a high-frequency cryogenic HEMT-amplifier, which was connected to a real-time spectrum analyzer (R&S FSW43, 512 MHz) set to IQ-analyzing mode.

III. RESULTS

The multiheterodyne RF spectrum is shown in Fig. 2(a). An acquisition time of 10 μs was used as a compromise between sufficient resolution bandwidth to resolve individual beat notes and short enough acquisition time to avoid significant spectral broadening due to the laser frequency instabilities. The major source of fluctuations, causing spectral overlap of the beat notes over longer timescales, are the instabilities in the offset frequencies of the lasers, which exceed those of the repetition rate by orders of magnitude. The figure shows twenty-two resolvable RF beat notes captured within the detector bandwidth, covering approximately 220 GHz in the optical domain.

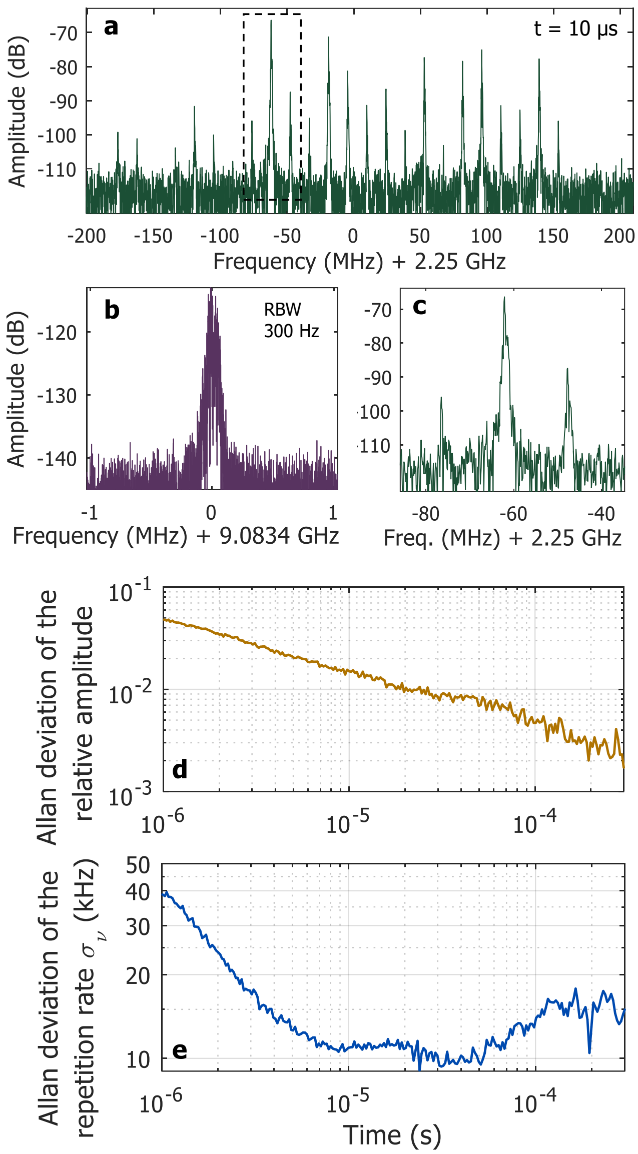

Fig. 2. (a) Multiheterodyne spectrum acquired within 10 μs showing twenty-two multiheterodyne beat notes, whose RF span corresponds to approximately 220 GHz in the optical domain. Despite the short acquisition time, the signal-to-noise of the majority of the beat notes exceeds 20 dB. **(b)** Intermode beat note extracted from the HEB through the high-frequency HEMT-amplifier. **(c)** Zoom of the strongest multiheterodyne beat note showing the increased noise pedestal. The neighboring weaker beat notes are also visible **(d)** Allan deviation plot of the instantaneous amplitude of the intermode beat note from (**b**), showing white-noise limited performance. **(e)** Allan deviation of the instantaneous frequency of the same signal indicating a drift after approximately 30 μs.

Figure 2(b) shows an example of an intermode beat note measured by the HEB. The beat note appears at ~9.1 GHz and can be tuned by ~15 MHz through either current or temperature. Further tuning disrupts the comb operation of the device and subsequently precludes multiheterodyne spectroscopy.

The intermode beat note amplitude and frequency stabilities are characterized by the Allan variance plots [6] shown in Fig. 2(c) and (d). The intermode beat note amplitude shows white-noise limited performance over the entire acquisition time, while its frequency, which corresponds to the FSR, start to drift at ~30 μs. This drift indicates the upper bound of the acquisition time used for coherent averaging unless real-time phase and timing correction to the multiheterodyne spectra can be implemented [7], [8].

IV. SUMMARY

In summary, we have studied the feasibility of multiheterodyne spectroscopy using terahertz QCL combs with integrated dispersion compensation, emitting sub-milliwatt of optical power. The multiheterodyne beat notes from two cryogenically-cooled THz-QCLs show frequency instabilities at the 30 μs time-scale, which hinders reliable retrieval of the spectroscopic information encoded in their amplitudes and phases. In order to perform accurate spectroscopy using these sources we are currently implementing mutually stabilizing locking circuitry and phase and timing correction procedures [8] that will enable coherent averaging [7], [9], [10] over extended time-scales. Spectroscopic results obtained with this system will be presented in details.

ACKNOWLEDGMENTS

The authors would like to acknowledge financial support by the DARPA SCOUT program (W31P4Q161001).

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