Terahertz dual comb spectroscopy using quantum cascade laser frequency combs

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Abstract: We demonstrate THz dual comb spectroscopy of molecular samples using dispersion compensated quantum cascade lasers. The system achieves an optical bandwidth of ~150 GHz at 2.9 THz and is used to measure ammonia in gas phase.

OCIS codes: (140.5965) Semiconductor lasers, quantum cascade; (300.6310) Spectroscopy, heterodyne; (300.6495) Spectroscopy, terahertz; (130.2035) Dispersion compensation devices

The terahertz (THz) spectral region (100 GHz – 30 THz) has gathered increasing interest in the spectroscopic community as technological advances in both generation and detection of THz radiation have provided new tools for selective identification of complex molecular structures in solid- ,liquid- , and gaseous phase. Generation of coherent THz radiation can be achieved in many ways, where the most prominent are based on photoconductive switches, optical rectification, difference frequency generation (DFG), or optical parametric oscillations (OPOs). However, none of these methods provide direct generation of THz radiation, which leads to added complexity of the spectrometers based on any of these techniques. Recent advances in the quantum cascade laser (QCL) technology [1] addresses this issue by enabling a chip-scale, monolithic semiconductor THz light source with milliwatts of optical power in the 1-5 THz range. Of particular interest is the 3-5 THz region, where the more established techniques, such as THz time-domain spectroscopy (TDS), suffer from poor signal-to-noise ratio due to a decrease in optical power at higher frequencies. Furthermore, THz-TDS systems often rely on mechanical moving optical delay lines, which necessitates acquisition times of seconds or longer and is prone to misalignment. Here, we propose an alternative by developing a THz system based on the QCL-based dual comb spectroscopy (DCS) technique [2,3] that has proven effective with lasers operating in the mid-infrared spectral domain.

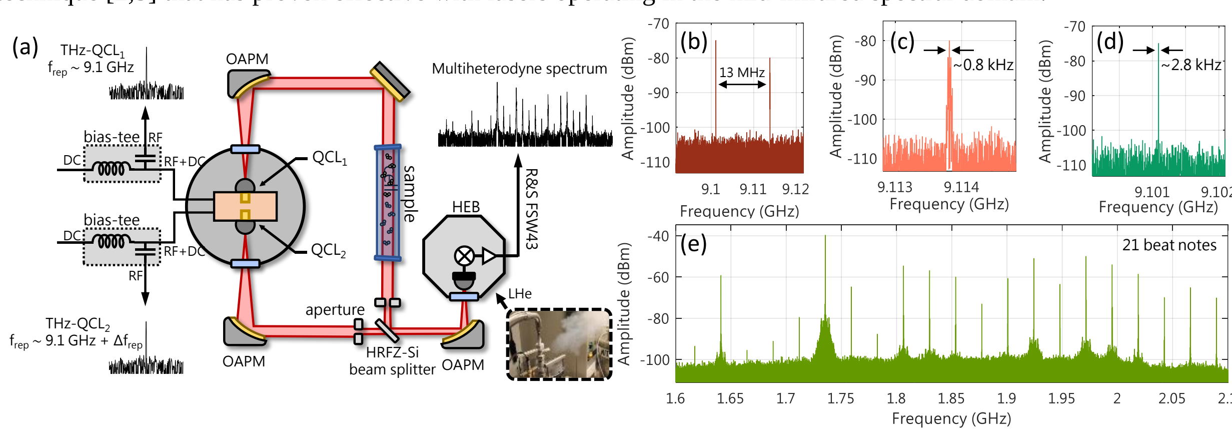

Fig. 1. (a) Schematic of the experimental setup. The Two THz-QCLs are facing opposite directions and their high frequency voltage characteristics are monitored by 12 GHz bias-tees. The THz radiation is detected by a high-bandwidth HEB (BW ~8 GHz). (b) Intermode beat notes as a result of mode self-beating measured at ~9 GHz. (c) Zoomed view of one of the intermode beat notes. (d) Zoomed view of the other intermode beat note. (e) Multiheterodyne beat note spectrum measured with 1 ms of acquisition time.

QCL-based THz-DCS presents additional technical challenges compared to its mid-infrared counterpart. First, contemporary THz-QCLs require cryogenic cooling during operation and with tens of watts of thermal load, a closed-system is preferred for extended periods of use. Unfortunately, even with careful attention to vibration dampening mechanisms, this generates a significant amount of mechanical vibrations, which adds noise to the system. Second, the rich THz spectrum of water vapor necessitates system-wide optical path purging to prevent complete extinction of the THz radiation. Third, high-performance THz-detectors rely on liquid helium cooling, which further increases the system complexity and reliance on cryogens. However, this field is rapidly progressing and room-temperature detectors based on Schottky-diodes are becoming increasingly available.

Despite these challenges we demonstrate the first THz dual comb measurements of a molecular sample in gas phase. A schematic of the experimental setup is given in Fig. 1(a). The THz radiation is generated by two dispersion compensated THz-QCLs [4] aligned in an anti-parallel configuration. Proper collimation is achieved by a combination of HRFZ-Si lenses attached directly to the laser, and 2-inch off-axis parabolic mirrors placed outside the cryostat. One of the two THz beams propagates through a custom-made absorption cell with wedged TPX windows (Tydex), after which it is spatially combined with the other beam by a HRFZ-Si beam splitter and focused on a hot electron bolometer (HEB, Scontel). The optical heterodyne mixing that occurs as a result of the mismatch of the free-spectral ranges (FSRs) of the THz-QCLs provide a direct link between the optical- and the radio-frequency (RF) domain. By this procedure the THz optical modes are mapped to the corresponding RF beat notes, which can be conveniently accessed by a real-time RF spectrum analyzer. Frequency comb operation is verified by measuring the optical self-heterodyne beating at the frequencies corresponding to the FSRs of the lasers. This information can also be obtained by measuring the RF characteristics of the voltage across the laser structures. In this case, a bias-tee is used to extract the high-frequency RF signal, while also supplying DC current to the lasers [see Fig. 1(a)]. The self-heterodyne beat notes for the two lasers are displayed in Fig. 1(b), (c), and (d), where (c) and (d) shows a 3 dB linewidth of ~0.8 kHz and ~2.8 kHz for the two beat notes, respectively. A typical RF spectrum containing 21 beat notes is shown in Fig. 1(e), which provides an instantaneous spectral coverage of ~190 GHz.

To demonstrate the spectroscopic capabilities of the system a measurement of high pressure ammonia sample was used. Although a simple molecule, such as ammonia, could be assessed more easily at other wavelengths, these measurements are a useful benchmark due to the availability of an accurate THz spectral model for comparison. An ammonia spectrum around ~3 THz using an acquisition time of 1 ms is shown in Fig. 2(a) together with a spectral model based on parameters from the HITRAN database. As can be seen the measurements agree well with the model whenever the beat notes exhibit sufficient carrier-to-noise ratio. A threshold for the beat note amplitude has been applied, which excludes the beat note at ~2.85 THz due to a large interference of water vapor. Figure 2(b) and (c) show the RF beat note spectra that was used to generate the spectrum of Fig. 2(a), where (b) is the reference measurement without absorber and (c) is the sample measurement. The attenuation due to ammonia is clearly visible on the right side of the figure.

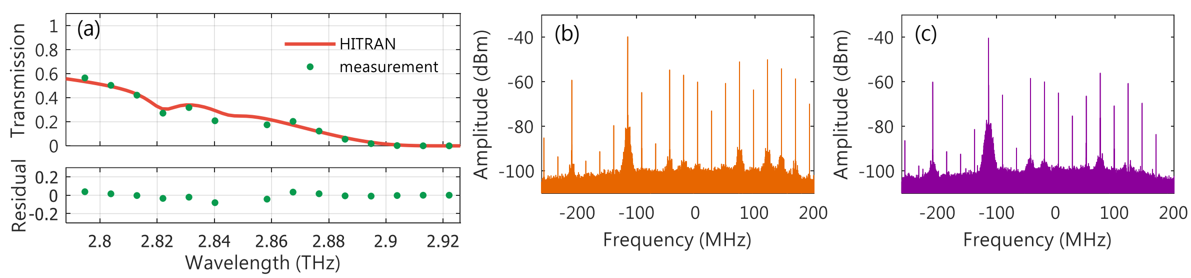

Fig. 2. (a) THz spectrum of 306 Torr of pure ammonia measured around 2.9 THz. A beat note amplitude threshold has been applied, which removes one data point from the spectrum due to significant interference from water vapor. (b) The reference RF beat note spectrum used for the measurement in (a). (c) The sample RF beat note spectrum used for the measurement in (a).

In summary, we demonstrate THz dual comb spectroscopy of ammonia using dispersion compensated THz-QCLs operating at ~3 THz. The system achieves an instantaneous spectral coverage of ~150 GHz and a short-term noise-equivalent absorption (NEA) of $<1\times10^{-3}/\sqrt{\text{Hz}}$. Efforts to increase the optical bandwidth of the light sources as well as further spectroscopic results and system performance characterization will be discussed in detail.

Acknowledgments

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