High-resolution dual-comb gas-phase spectroscopy with a mode-locked laser on a photonic chip

Z. Chen,[1] K. Van Gasse,[2] E. Vicentini,[1,3,4] J. Huh,[1] S. Poelman,[2] Z. Wang,[2] G. Roelkens,[2] T.W. Hänsch,[1,5] B. Kuyken,[2] N. Picqué [1]

[1]*Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Straße 1, 85748, Garching, Germany*
[2]*Photonics Research Group, INTEC, Ghent University - imec, Technologiepark-Zwijnaarde 126, 9052 Ghent, Belgium*
[3]*Dipartimento di Fisica, Politecnico di Milano, Piazza L. Da Vinci, 32 20133 Milano, Italy*
[4]*Istituto di Fotonica e Nanotecnologie, Consiglio Nazionale delle Ricerche, Piazza L. Da Vinci, 32 20133 Milano, Italy*
[5]*Ludwig-Maximilians-Universität München, Fakultät für Physik, Schellingstr. 4/III, 80799, München, Germany*
zaijun.chen@mpq.mpg.de

Abstract: An integrated III-V-on-silicon mode-locked laser enables the first on-chip comb generator of 1.0-GHz line-spacing for direct interrogation of gas-phase narrow rovibrational transitions in molecules. Its flat-top spectrum empowers real-time multiplexed spectroscopy without any scanning elements.

Laser frequency combs enable novel approaches to molecular spectroscopy over broad spectral bandwidths [1]. Progress towards integrated frequency-comb sources shows an attractive potential of broadband rapid spectroscopy with ultra-miniaturized devices [2]. Several platforms of chip-based comb generators, including Kerr combs [3], electro-optic microrings [4] and quantum cascade lasers [5], have led to proof-of-principle demonstration of gas-phase spectroscopy. Unfortunately, these comb sources feature a large comb-line spacing, on the order of ten GHz. The width of a molecular transition at atmospheric pressure is usually of a few GHz. The existing demonstrations harnessing on-chip combs have not allowed to properly sample the molecular profiles without any tuning of the comb lines. The intensity variations between comb lines and the small number of usable comb lines have also limited the sensitivity and the spectral bandwidth.

Here we explore integrated mode-locked laser comb sources for broadband high-resolution gas-phase spectroscopy. They are based on III-V-on-silicon mode-locked lasers with 1-GHz comb line spacing [6], which is sufficient to sample absorption profiles of molecules in gas-phase. The performance of one such a source for spectroscopy is assessed in a dual-comb multi-heterodyne scheme [1]. We beat the III-V-on-silicon mode-locked laser with an electro-optic (EO) frequency comb synthesizer of slightly different repetition frequency. Then we perform dual-comb spectroscopy of carbon monoxide (CO) at atmospheric pressure with 1-GHz resolution.

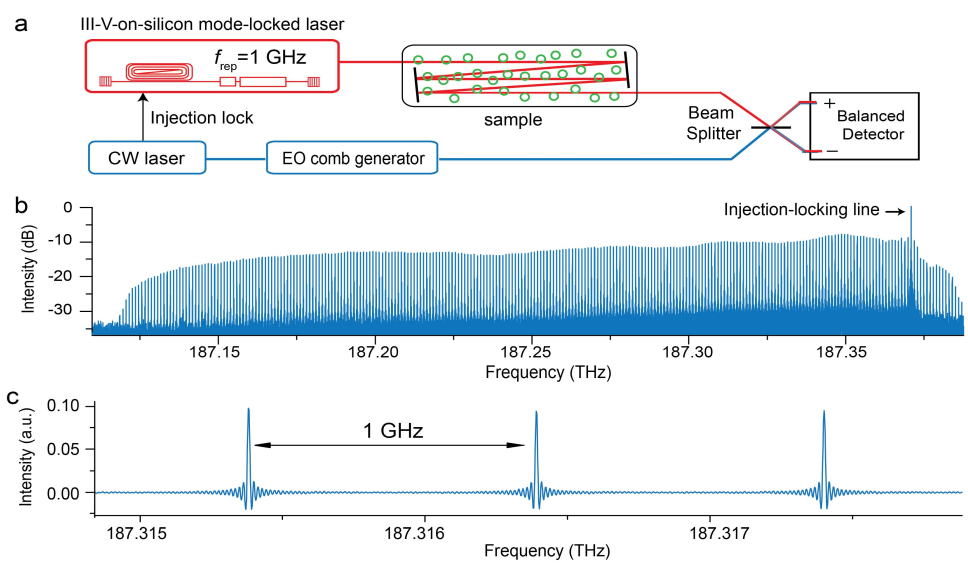

Figure 1: High-resolution dual-comb spectroscopy with a III-V-on-silicon mode-locked laser. (a) Experimental setup. (b) apodized dual-comb spectrum with resolved comb lines. The spectrum is recorded in the radio-frequency domain and the frequency scale is rescaled to the optical domain. The intense comb line is the beat note between the injection-locking comb mode and the carrier of the EO comb at 187.38 THz. (c) a magnified view of (b) with three unapodized comb lines of 1-GHz spacing. CW: continuous-wave. EO: electro-optic

The experimental setup is sketched in Fig. 1a. The integrated mode-locked laser was fabricated by die-to-wafer bonding of a III-V material (InP/InGaAsP 6 QWs) on top of a silicon-on-insulator photonic chip (400 nm thick active Si layer) followed by several wet and dry etching steps [6]. The laser consists of an amplifier with a length of 0.7 mm and a passive silicon waveguide cavity of 38 mm in an anti-colliding pulse topology. The laser consumes only 140 mW of electrical power. It emits around 187 THz with a pulse repetition rate of f_{rep}=1.0096 GHz. Its spectrum is flat-top over a 3-dB span of 1.17 THz with 1154 comb lines. The output of the laser is amplified and sent to a multipass cell with an absorption path of 76 m. A continuous-wave (CW) laser of narrow linewidth is injected into the mode-locked laser cavity. The same CW laser seeds the EO comb generator [7] of a repetition frequency of $f_{rep}+\Delta f_{rep}$ (Δf_{rep}=1 MHz) and it thus provides mutual coherence in the dual-comb interferometer. The beam of the mode-locked laser and that of the EO comb are superimposed with a beamsplitter. At the output of the beamsplitter, a balanced detector records the time-domain interference between the two combs. The dual-comb radio-frequency spectrum is revealed with a Fourier transform of the interferogram. The frequency scale is converted to the optical domain.

An apodized spectrum centered around 187.25 THz (Fig. 1b) is recorded within 15 ms. It results from 150 averages of spectra of 100 µs each. The spectrum is smooth and flat-top over a span of 262 GHz with 260 resolved comb lines. Such a bandwidth is limited by the coverage of the EO comb but the III-V-on-silicon laser allows for a span of 1.17 THz. A magnified representation of three unapodized comb lines spaced by 1.0096 GHz (Fig. 1c) shows that the lines appear with a cardinal-sine line shape, resulting from the Fourier transform of an interferogram of finite measurement time. This shows that the injection-locking scheme satisfactorily establishes coherence between the two interfering combs.

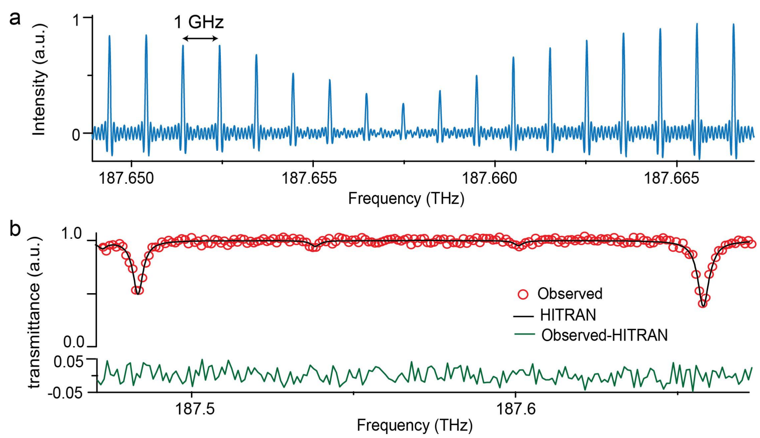

Figure 2: Experimental absorption spectra of CO. (a) The P(19) line of the 3-0 band of ^{12}CO sampled by the comb lines. (b) A portion of a transmittance spectrum sampled at the maximum of each comb line with a resolution of 1 GHz.

The gas cell is filled with CO in natural abundance with a pressure of 9.87×10^4 Pa. The P(19) line of the 3-0 band of ^{12}CO has a collisional linewidth of 3.3 GHz, which results in a full-width at half-maximum of 4.6 GHz in the transmittance profile. The profile is satisfactorily sampled by the comb lines of 1-GHz spacing (Fig.2a). A transmittance spectrum (portion shown in Fig. 2b) is measured within 200 µs. The signal-to-noise ratio over the entire span of 0.4 THz is 52, corresponding to 3700 $s^{-1/2}$. The two strong molecular lines are the P(20) and P(19) lines of 3-0 band of ^{12}CO and the weak lines are the R(13)-R(15) lines in ^{13}CO. The experimental spectrum agrees with a discrepancy of 2% with a spectrum calculated using the line parameters available on HITRAN 2016.

Our proof-of-principle demonstrates the potential of III-V-on-silicon comb generators for real-time high-resolution multiplexed spectroscopy. It represents a significant milestone towards fully-integrated spectrometers manufacturable at the wafer scale.

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