

# Dual comb operation of $\lambda \sim 8.2 \mu\text{m}$ Quantum Cascade Laser frequency comb with 1 W optical power

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In this work, we report the characterization of a quantum cascade laser frequency comb with 1.05 W optical power at  $\lambda \sim 8.2 \mu\text{m}$ . The 4.5 mm long device, has a high reflectivity coating on the back facet as well as a top cladding designed to lower the group velocity dispersion and is operated at 258 K. Very strong (more than 60 dB) narrow beatnotes are shown and frequency comb operation is obtained on a bandwidth of  $85 \text{ cm}^{-1}$  in a very large range of the light-versus current characteristics. A bandwidth of  $82 \text{ cm}^{-1}$  has a power per mode of more than 1 mW and an average power per mode of 4.1 mW. Finally, a multi-heterodyne spectrum with 215 lines covering an optical bandwidth of more than  $70 \text{ cm}^{-1}$  measured with lasers showing similar performances is presented with very good line separation.

There is a strong interest in the development of optical frequency combs in the mid-infrared as the implementation of dual-comb spectrometers<sup>1,2</sup> in this frequency range that exhibits strong fundamental lines of chemicals could revolutionize chemical sensing as well as high resolution spectroscopy. As compared to other approaches to mid-infrared comb generation<sup>3</sup>, the quantum cascade lasers (QCL) have the attractive features that they are powerful, compact and electrically driven. In these devices, frequency comb operation can be achieved thanks to four wave mixing<sup>4</sup> and dual-comb spectroscopy has already been demonstrated<sup>5,6,7</sup>. In order to further improve the sensitivity and the acquisition speed of dual-comb spectrometers, significant efforts have been dedicated both to the increase of the optical power output<sup>8</sup> and the dynamical range for which the QCL operates as a frequency comb<sup>9,10</sup> in order to take advantage of the maximum output power and bandwidth coverage of the laser.

Even if QCL frequency combs are stable under operation, they are sensitive to small changes in optical feedback. It is therefore important to do all characterizations under the same optical conditions. To ensure consistent data characterization, we adopted the setup schematized in FIG. 1. a). The laser is driven with a low noise driver and a bias-T sends the RF part of the current on a spectrum analyzer to characterize the frequency comb beatnote. The beam is collimated with a high numerical aperture lens of  $\text{NA}=0.85$ . In order to isolate the laser from back-reflections from the Fourier Transform Infrared Spectrometer

(FTIR), a tilted neutral density filter (NDF) with 1% transmission is placed after the lens. A beam splitter can be used to monitor the evolution of the optical power after the NDF (see appendix A). Finally, the spectra are measured with a FTIR with a resolution of  $0.075 \text{ cm}^{-1}$  and the optical power is measured by placing a thermopile sensor directly after the lens. In this configuration, we verified that no element placed after the NDF will disturb the beatnote measured on the spectrum analyzer. FIG. 1.(b) shows two spectra obtained under the same driving conditions but once with a narrow beatnote and once with a broad beatnote. The broad beatnote was obtained by placing the NDF perpendicular to the beam to increase the optical feedback in the laser and destabilize the frequency comb operation. The two spectra have a different bandwidth; it is reduced by more than  $15 \text{ cm}^{-1}$  when in frequency comb operation. It is thus crucial to do all optical spectra measurements of the frequency comb while conserving the narrow beatnote properties during the measurement. All measurements presented here were done using this method. In some cases nevertheless, weak optical feedback can be of use such as in <sup>11</sup> where it allowed to generate self-mixing effect to confirm comb operation.

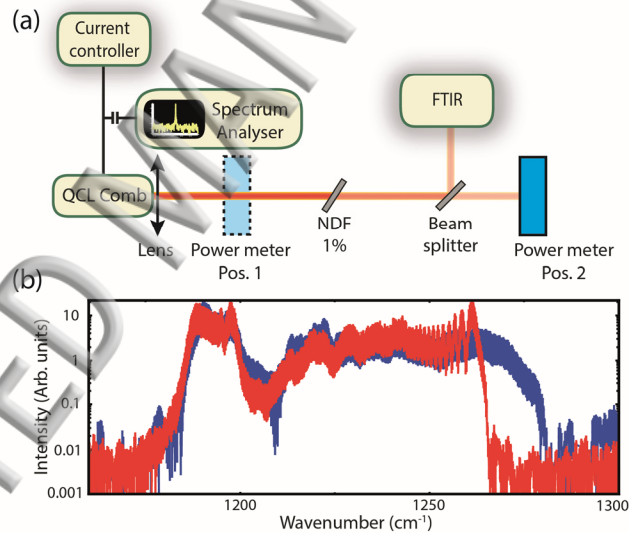


FIG. 1. a) Schematic of the frequency comb characterization setup. b) Optical spectra measured with (blue curve) and without (red curve) feedback i.e. with broad and narrow beatnote, respectively.

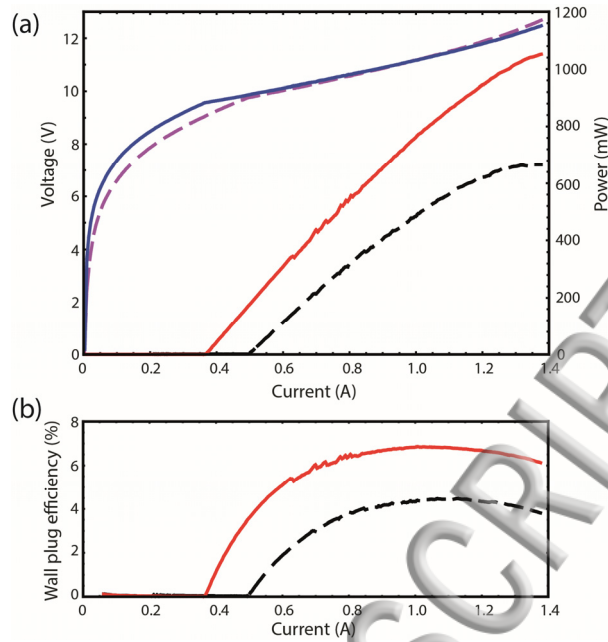


FIG. 2. a) LIV characteristic of the 4.5 mm long laser with HR coating at 258 K (full lines) and 293 K (dashed lines). b) Corresponding wall plug efficiency.

The QCL active region is a strain compensated InGaAs/AlInAs dual stack heterostructure grown by MBE (see appendix B). The laser ridge is 4.5 mm long and has a high reflectivity (HR) coating on the back facet ( $\text{Al}_2\text{O}_3/\text{Au}$ ). The Light-Current-Voltage (LIV) characteristics and the quantum efficiencies are presented in FIG. 2 for temperatures of 258 K and 293 K. Powers above 1 W and efficiency of 6.85% are reached at 258 K. Subthreshold spectra measurements were performed at 293 K using a nitrogen cooled mercury-cadmium-telluride detector. The deduced group velocity dispersion (GVD) and modal gain <sup>12</sup> are plotted in FIG. 3. The top cladding of the laser has been specially designed following the approach described in <sup>13</sup> to obtain a low GVD to favor frequency comb operation. From simulations, it lowers the group delay dispersion (GDD) of the laser by 1800  $\text{fs}^2$  in average compared to a very thick cladding. For this device, the GDD measured below threshold is between -2000  $\text{fs}^2$  and -7000  $\text{fs}^2$  in the emission bandwidth.

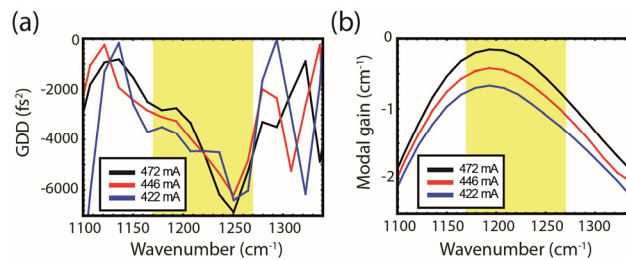


FIG. 3 a) Measured GDD at different driving currents at 293 K. b) Measured modal gain for different driving currents at 293 K. The shaded areas show the bandwidth on which the laser operates.

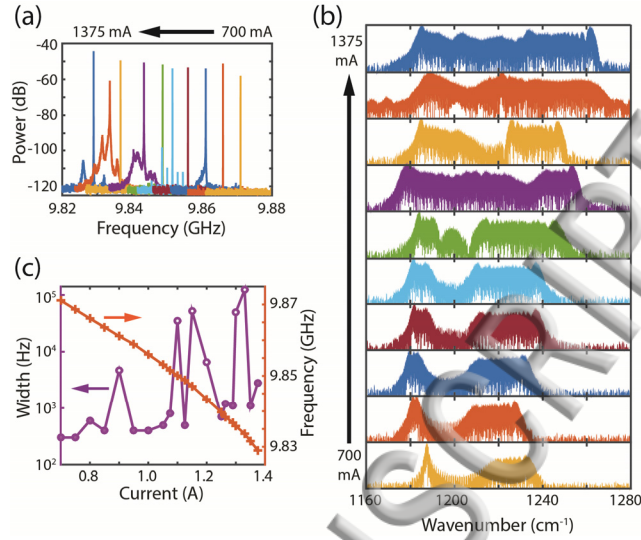


FIG. 4. a) Measured beatnote at 258 K with a span of 20 MHz and a bandwidth resolution of 100 Hz for driving currents of 700, 800, 900, 1000, 1075, 1125, 1200, 1290, 1330 and 1375 mA. b) Corresponding optical spectra. c) Beatnote width at -20 dB and frequency for the different currents measured at 258 K.

At 258 K, different beatnote types were recorded across the dynamical range (the laser becomes multimode shortly before 700 mA) and are shown in FIG. 4 a). It is possible to observe those different types mostly because of the extremely high signal to noise ratio (SNR) obtained thanks to very strong beatnote signals (up to more than 60 dB). We attribute the strong SNR to the fact that the intra-cavity power is very high and therefore a strong modulation of the current is induced in the laser. For most currents, a sharp beatnote was observed (showing sometimes sub-lines like for 1075 or 1375 mA but with intensities more than 40 dB below the main peak); for some currents, a pedestal was visible (at 900 and 1200 mA for example) and finally some other currents gave broad and multi peaks beatnote (at 1330 mA for example). These observations show that many different regimes can exist in a QCL and that a careful characterization has to be conducted before using them for spectroscopy purpose for example. The corresponding optical spectra measured using a Deuterated triglycine sulfate detector are shown in FIG. 4 b). For spectroscopy purpose, it is more meaningful to characterize the quality of the beatnote with its width at -20 dB instead of -3 dB as usually presented in literature; indeed, a too high width at -20 dB would mean a merging of neighboring lines in a dual comb multi-heterodyne signal. The beatnote width at -20 dB and the beatnote frequency as a function of current are shown in FIG. 4 c); for the beatnote width, the full circles show the frequency comb operation while the empty circles show the high phase noise regime operation (width above several hundreds of kHz). A close up on the beatnote using a span of 200

kHz and a bandwidth resolution of 100 Hz is shown in FIG. 5 a) for a current of 1375 mA at 258 K; a sharp and strong (SNR > 60 dB) beatnote is visible, signature of frequency comb operation. Neglecting dispersion and because of the similar dependence of the effective and group index on external parameters, the ratio of the optical to beatnotes linewidth is given roughly by the order of the line, in our case  $N=3740$ . Therefore, the 3kHz beatnote linewidth measured at -20dB would correspond to an optical linewidth at -20dB of 11MHz which corresponds approximately to  $0.0003 \text{ cm}^{-1}$ . The mode spacing and power per mode are shown in FIG. 5 b) for spectra taken at 258 K and at room temperature for a current of 1375 mA. At 258 K, a total bandwidth of  $85 \text{ cm}^{-1}$  with a power of 1.05 W is achieved and a continuous bandwidth of  $82 \text{ cm}^{-1}$  has a power per mode above 1 mW with an average power of 4.1 mW. At room temperature, a total bandwidth of  $75 \text{ cm}^{-1}$  with a power of 0.664 W is achieved and a continuous bandwidth of  $73 \text{ cm}^{-1}$  (excluding 5 modes for wavenumbers in the vicinity of  $1225 \text{ cm}^{-1}$ ) has a power per mode above 1 mW with an average power of 2.99 mW. At room temperature, the beatnote is more robust and is a narrow peak on all the dynamical range (the laser becomes multimode shortly before 900 mA) beside at 1100 mA (see FIG. 5 c)). Those values are similar to Ref. 9 in terms of frequency comb power at room temperature but here the laser is able to operate in frequency comb regime up to its roll-over thanks to its low GVD.

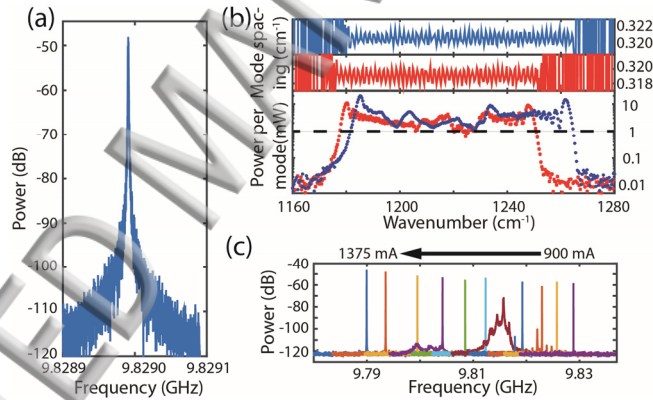


FIG. 5. a) Measured beatnote at 1375 mA and 258 K with a span of 200 kHz and a bandwidth resolution of 100 Hz. b) Neighbor mode spacing at 258 K and at 293 K (top and middle panel, respectively) and power per mode (lower panel) for a current of 1375 mA at 258 K and 293 K (blue and red points, respectively). c) Measured beatnote at 293 K with a span of 20 MHz and a bandwidth resolution of 100 Hz for driving currents of 900, 950, 1000, 1050, 1100, 1150, 1200, 1250, 1300, 1350 and 1375 mA.

Following those results, two QCL frequency combs from the same processed epi-layer which show similar performances have been used as sources in a commercial dual-comb spectrometer<sup>6</sup>. The multi-heterodyne beatnote obtained for a single shot measurement with 20 ms integration time is shown in FIG. 6. The shape of this multi-heterodyne spectra results from the convolution of the optical spectra of both laser as well as from optical properties of the setup such as optical elements

transmission spectra or beam overlap on the detector. The data processing was done following the approach described in the supplementary material of <sup>5</sup>. The lasers are 4.5 mm long and are operated one at 273 K and the other at 288 K with currents of approximately 1270 and 1386 mA respectively without further stabilization. Even if uncoated, each laser has an optical output power above 400 mW in those driving conditions. About 215 peaks corresponding to an optical bandwidth of more than  $70\text{ cm}^{-1}$  are observable between 200 and 600 MHz with a spacing of approximately 1.77 MHz. This corresponds to an increase of bandwidth coverage by a factor of at least 1.5 from previous published works <sup>5,7,13,14</sup>. The inset shows a zoom on a few modes to pinpoint the good separation of each lines of the multi-heterodyne spectra, which is of paramount importance for quantitative spectroscopy measurements. The width of each line is of few hundreds of kHz due to the drifts of the comb during the 20ms acquisition time. Spectroscopy measurements have been done with another pair of lasers from the same process in <sup>15</sup>. For 100  $\mu\text{s}$  integration time, amplitude variations of a single emission line of 2 permille rms were observed.

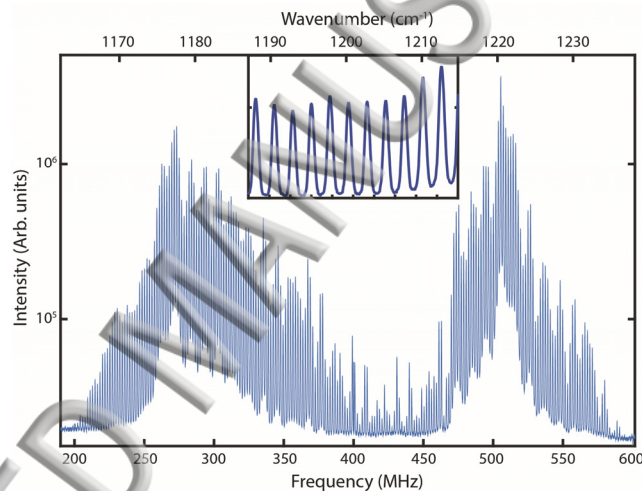


FIG. 6. Multi-heterodyne spectra for an integration time of 20 ms and zoom in on a few lines between 230 and 250 MHz (inset). The wavenumber scale is deduced from earlier characterization of the lasers and have an uncertainty on the central frequency of  $\pm 5\text{ cm}^{-1}$ .

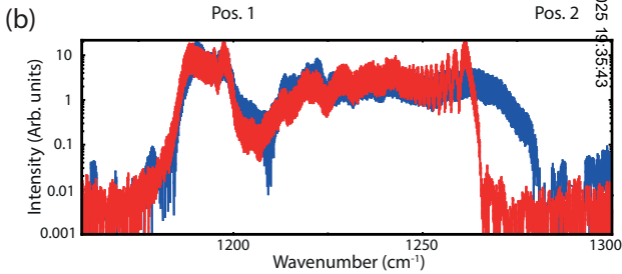
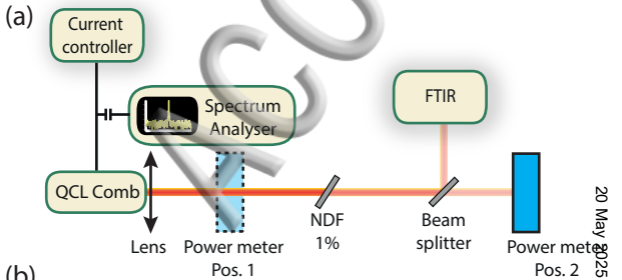
In summary, we demonstrated above 1 W power QCL frequency comb emitting around  $8.2\text{ }\mu\text{m}$ . More than 270 modes covering a bandwidth of  $85\text{ cm}^{-1}$  and a continuous bandwidth of  $82\text{ cm}^{-1}$  showing a power per mode above 1 mW at 258 K has been obtained. At room temperature, the QCL operates in the frequency comb regime on almost the full dynamical range giving up to 664 mW output power with similar bandwidth. Due to their particularly interesting properties for spectroscopy measurements, lasers from the same process showing similar performances have already been integrated by partners in dual-comb spectroscopy setups. In particular, we present a broad multi-heterodyne spectra with 215 lines covering an optical bandwidth of more than  $70\text{ cm}^{-1}$ .

This material is based upon work supported by the Defense Advanced Research Projects Agency (DARPA) Program Office under Contract No. W31P4Q-15-C-0083 and the Schweizerischer Nationalfonds zur Förderung der Wissenschaftlichen Forschung (SNF) under Contract No. 200020\_165639.

### Supplementary material:

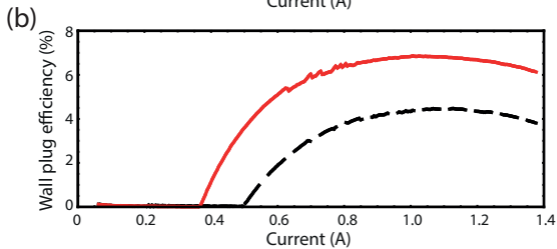
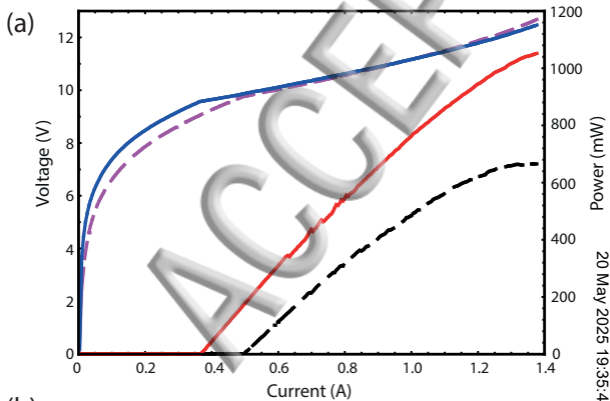
In the first part of the supplementary material we demonstrate that under the same driving conditions the optical power stays the same when operating in the frequency comb regime or in the broad beatnote regime. In the second part we give the layer sequence of the QCL presented here.

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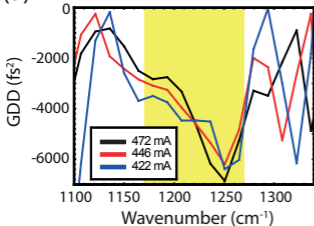


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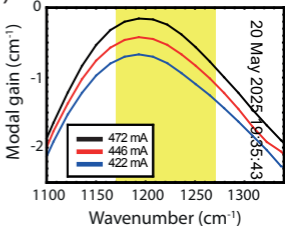


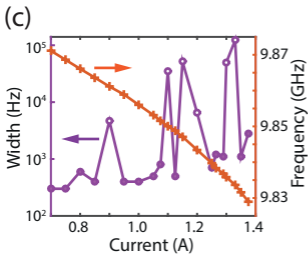
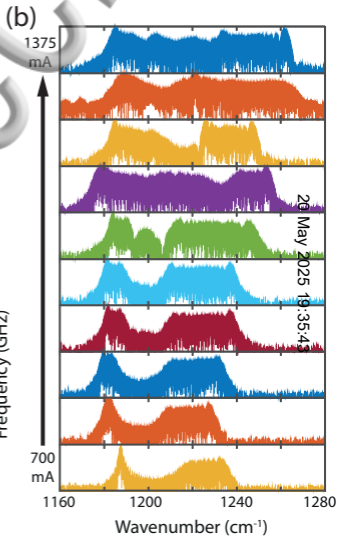
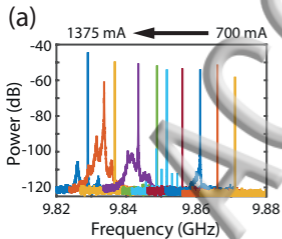


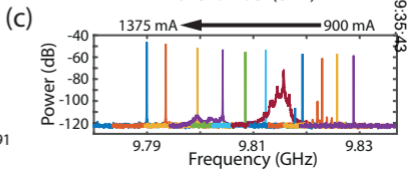
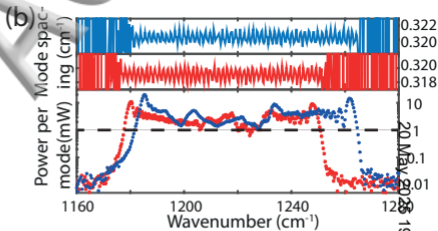
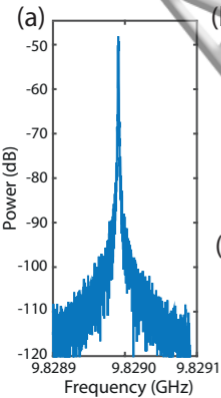
(a)



(b)







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