Dispersion engineering of quantum cascade laser frequency combs: supplementary material

GUSTAVO VILLARES^{1,*}, SABINE RIEDI¹, JOHANNA WOLF¹, DMITRY KAZAKOV¹, MARTIN J. SÜESS¹, PIERRE JOUY¹, MATTIAS BECK¹, AND JÉRÔME FAIST^{1,†}

¹Institute for Quantum Electronics, ETH Zurich, 8093 Zürich, Switzerland

*Corresponding author: gustavo.villares@phys.ethz.ch

[†]e-mail: jfaist@phys.ethz.ch

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1. QCL-COMB CHARACTERIZATION

The performance of QCL-combs is characterized by simultaneously acquiring the RF spectrum containing the comb repetition frequency and the optical spectrum. The QCL-combs are driven with low noise current drivers (Wavelength electronics QCL500 OEM or QCL2000 LAB) with a specified average current noise density of $2 \text{ nA}/\sqrt{\text{Hz}}$. The temperature fluctuations of the lasers were also reduced to less than 10 mK by using a low thermal drift temperature controller (wavelength electronics PTC10K-CH) with a $50 \text{ k}\Omega$ thermistor. The comb repetition frequency is measured through the RF port of a bias-tee inserted between the current driver and the QCL-comb (c.f. Fig. 1a of the main text) [1, 2]. A RF spectrum analyzer (Rhode & Schwarz FSU50) is used to acquire the RF spectrum. After collimation by a high-numerical aperture (0.86) aspheric lens, the QCL-comb output is sent to a FTIR (0.12 cm^{-1} resolution) in order to acquire the optical spectrum (c.f. Fig. 1a of the main text).

2. INTERMODE BEAT SPECTROSCOPY

We have shown in the main text how intermode beat spectroscopy has been used for investigating the comb in the high-phase noise regime, in order to investigate if one spectral region of the comb spectrum is mainly responsible for the comb destabilization. We report here additional information on the device used for this technique, as well as additional data on the intermode beat spectroscopy characterization of this device. The device used is a 6 mm long QCL that comes from the same laser process as all device shown in this works, and is HR coated on the back facet. Fig. S1a and Fig. S1b show respectively the optical and RF spectrum of this device. As already discussed on

the main text, both comb regimes and high-phase noise regimes are observed. Fig. S1c shows the intermode beat interferogram, obtained by setting the FTIR in step-scan mode and acquiring the RF spectrum of the photocurrent generated when using a liquid nitrogen cooled quantum well infrared detector (QWIP) as an external detector of the FTIR. The Fourier transform of this interferogram gives the intermode beat spectrogram shown in Fig. 1 of the main text.

3. DISPERSION CHARACTERIZATION OF QCL-COMBS

We describe in this section the procedure to measure the dispersion introduced by GTI mirrors as well as the dispersion of a QCL-comb source. We first describe the entire procedure for the case of GTI mirrors. The method employed for the dispersion measurement of the GTI structures and of the QCL-comb sources being very similar, only their differences will be detailed.

For each evaporation of a GTI mirror structure, an InP substrate ($320 \mu\text{m}$ thick) is placed in the evaporation chamber and is used as a reference sample. This reference sample is used to characterize the dispersion introduced by the GTI mirror. The characterization is done by placing the device under test (DUT) after the beam-splitter of a FTIR, where the beams are recombined, as shown schematically on Fig. 2c of the main text. An interferogram generated by the reflection upon the GTI mirror is then acquired. A typical interferogram is shown in Fig. S2a, where we observe an intense center burst, corresponding to the position of the moving mirror where both arms are introducing the same optical delay. More importantly, several small satellites are also observed, indicated by the red dots on Fig. S2a. As explained schematically on Fig. 2b of the main text, the GTI layer is deposited on top of an InP substrate for its characterization and

Fig. S1. intermode beat spectroscopy (additional data). **a** Optical spectra acquired at $T = -15^\circ\text{C}$ for different current values and measured with a FTIR (0.12 cm^{-1} resolution) of a QCL-comb (6.0 mm long, standard HR coating on the back facet, episcide-down mounted on AlN submount) in CW operation. **b** Electrical RF spectra acquired at $T = -15^\circ\text{C}$ at the same values of current as in Fig. S1a, measured with a spectrum analyser (span = 50 MHz, resolution bandwidth (RBW) = 30 kHz, acquisition time = 20 ms). The RF spectra are centered at 7.74 GHz, corresponding to the RF beatnote created by a 6.0 mm long device. Comb and high-phase noise regimes are highlighted. **c** Intermode beat interferogram acquired by setting the FTIR in step-scan mode (0.25 cm^{-1} resolution) and acquiring the RF spectrum (span = 200 MHz, RBW = 100 kHz, acquisition time = 200 ms) of the photocurrent generated when using a QWIP (> 30 GHz bandwidth) as an external detector of the FTIR.

the structure is illuminated through the substrate. Therefore, the satellites originate from the interference between the front surface of the InP substrate and the one occurring after one or more round-trips in the InP substrate combined with the GTI structure, which will thus incur the phase shift due to the GTI coating. The first and most intense satellite (position 14000 in Fig. S2a) corresponds to the case of a single roundtrip and contains the information concerning the dispersion introduced by the GTI. In order to calculate the dispersion introduced by the GTI, the first satellite is numerically isolated and apodized, as shown in Fig. S2b. The length of this interferogram has to be carefully chosen, as it determines the spectral resolution as well as the accuracy of the GDD measurement [3]. After apodization, we perform a fourier transform (Fast Fourier Transform algorithm) and compute the phase spectrum, which is shown in Fig. S2c. This phase corresponds to the accumulated phase when light experiences a roundtrip inside the substrate and GTI mirror, as shown schematically in Fig. 2b of the main text. Finally, as the

group delay dispersion (GDD) is defined as

$$\Delta\phi(\omega) = \underbrace{\Delta\phi_0}_{\text{Phase at } \omega_0} + (\omega - \omega_0) \underbrace{\left(\frac{d\Delta\phi}{d\omega}\right)_{\omega_0}}_{\text{Group Delay}} + \frac{1}{2}(\omega - \omega_0)^2 \underbrace{\left(\frac{d^2\Delta\phi}{d\omega^2}\right)_{\omega_0}}_{\text{GDD}} \quad (1)$$

the GDD can be obtained by computing the second derivative of the relative phase $\Delta\phi$ according to ω . The measured GDD is displayed on Fig. 2c of the main text.

Fig. S2. Method for dispersion characterization of GTI mirrors as well as QCL-combs. **a** Interferogram generated by a FTIR when measuring the reflection of a GTI mirror. The red dots correspond to the different satellites observed on the interferogram. The resolution is set to 1.7 cm^{-1} , enough to be able to observe up to the second satellite created on the interferogram. **b** Zoom on the first satellite, where the dispersion information is contained. The number of points of this isolated interferogram sets the frequency resolution of the GDD measurement as well as the GDD accuracy. **c** Relative phase spectrum extracted from the first satellite of the interferogram. **d** Reflectivity of the different coatings (HR and GTI) used in this work. The GTI structure characterized here corresponds to the GTI introducing negative GDD, detailed on Table S1. The reflectivity of the GTI drops on the spectral region below 1300 cm^{-1} , due to the absorption of SiO_2 . The comb spectral region is also highlighted.

Finally, we described the method used for characterizing the dispersion of MIR QCL-combs. The QCL-comb is driven under threshold and is aligned to a FTIR, which is used to acquire the interferogram generated by the sub-threshold radiation. The first satellite observed in the interferogram corresponds to the relative phase between the light being directly emitted by one facet and the light experiencing a roundtrip into the cavity and subsequently emitted by this same facet [4]. Therefore, the information concerning the dispersion of a QCL-comb is contained into this part of the interferogram. By applying the same numerical method described for the analysis of the dispersion introduced by GTI mirrors, the phase spectrum as well as the

GDD of a QCL-comb can be computed. The GDD of a QCL-comb coated with a GTI mirror is shown in Fig.3 c of the main text.

4. GTI MIRRORS FOR DISPERSION COMPENSATION

The GTI mirrors designed for our QCL-combs consist of different layers of dielectric materials and a final layer of gold. Ideally, the layers of the GTI have to be transparent or at least introduce negligible absorption. We therefore used a combination of Al_2O_3 and SiO_2 , as they are relatively transparent in the spectral region of the QCL-combs designed for this study, as discussed below. The operation of the GTI mirror is done such that either a minimum or maximum of the GDD introduced by the mirror lies in the comb spectral region, in order to introduce negative or positive dispersion, respectively. The design is performed numerically by using a simulation tool computing the complex reflectivity of the coating, based on a transfer matrix formalism. However, the starting point of the design is found by using the analytical expression of the GDD introduced by a GTI made with a perfect transparent material

$$\text{GDD}_{\text{GTI}} = -\frac{2\tau_0^2(1 - R_t)\sqrt{R_t}\sin(\omega\tau_0)}{(1 + R_t - 2\sqrt{R_t}\cos(\omega\tau_0))^2} \quad (2)$$

where τ_0 is the cavity roundtrip time, R_t is the reflection coefficient of the low reflectivity facet of the GTI and ω is the optical frequency. We used the transfer matrix simulation tool only to improve and refine this initial design, in which the layered structure and the absorption of the different layers were not considered. The precision of the individual layer thicknesses is of critical importance for the correction of the dispersion introduced by the GTI mirror. To satisfy the required precision, calibration runs were conducted before every GTI evaporation to ensure variations of the thicknesses on the order of 3% percent.

Table S1. Detailed structure of GTI mirrors developed for dispersion compensation of QCL-combs. The first evaporated layer correspond to the first layer of the table.

GTI for positive GDD		GTI for negative GDD	
Material	Thickness	Material	Thickness
Al_2O_3	250 nm	Al_2O_3	511 nm
SiO_2	250 nm	SiO_2	102 nm
Al_2O_3	250 nm	Al_2O_3	511 nm
SiO_2	250 nm	SiO_2	102 nm
Al_2O_3	250 nm	Al_2O_3	511 nm
SiO_2	250 nm	SiO_2	102 nm
Al_2O_3	250 nm	Al_2O_3	511 nm
SiO_2	250 nm	SiO_2	102 nm
Al_2O_3	250 nm	Al_2O_3	511 nm
SiO_2	250 nm	SiO_2	102 nm
Al_2O_3	150 nm	Al_2O_3	154 nm
Au	150 nm	Au	154 nm

Fig. S3. Dispersion compensated QCL-combs (additional data). **a** Power-current-voltage of a QCL-comb (4.5 mm long, episcide-down mounted on AlN submount) coated with a GTI mirror introducing negative dispersion. The measurements are done in CW operation at different temperatures. Single-mode and Comb regimes are highlighted. **b** Electrical RF spectra acquired at $T = -16^\circ\text{C}$ at different values of current, measured with a spectrum analyser (span = 50 MHz, RBW = 300 kHz, acquisition time = 20 ms). The RF spectra are centered at 9.814 GHz, corresponding to the RF beatnote created by a 4.5 mm long device. The measured RF spectra show single and narrow beatnotes (FWHM < 30 kHz). No high-phase noise regime is observed. **c** Optical spectra acquired at $T = -16^\circ\text{C}$ at the same values of current as in Fig. S3b and acquired with a FTIR (0.12 cm^{-1} resolution). The QCL-comb spectrum is centered at 1360 cm^{-1} and spans over 50 cm^{-1} in the comb regime.

We report now the different designs of GTI mirrors that were used to introduce positive/negative values of dispersion, as described in the main text. Table S1 shows the detailed structure of the different designs of GTI mirrors. The GTI mirrors are composed of different layers of Al_2O_3 and SiO_2 and are always terminated with a thin layer of gold. As discussed in the main text, a discrepancy between the simulated values of the GDD introduced by a GTI mirror and the measured values of the GDD is observed around 1270 cm^{-1} . This is explained by the absorption introduced by SiO_2 . Fig. S2d shows the reflectivity of a GTI mirror containing Al_2O_3 and SiO_2 layers as well the spectral region where the comb operates. The fringes observed on both

measurements arise from the interferences between the reflection from the front surface of the InP substrate and the coating. Although Al_2O_3 can be assumed perfectly transparent in the comb operation region, SiO_2 is slightly absorbing in this spectral region, responsible for the increase of the absorption of the GTI structure for wavenumbers below 1315 cm^{-1} , as observed on Fig. S2d. This small absorption introduces an additional term to the relative phase $\Delta\phi$ introduced by the GTI, which directly translates into an additional source of dispersion. By using this additional source of GDD, we were able to introduce around -7000 fs^2 at 1270 cm^{-1} , a value of GDD that was not possible to achieve with a totally transparent GTI mirror with the same thickness.

5. ADDITIONAL QCL-COMB WITH NEGATIVE GTI MIRROR

In this section, we show additional data concerning QCL-combs coated with GTI mirrors introducing negative dispersion. We fabricated several devices with the same length from the same laser fabrication process. The same design of GTI mirror introducing negative dispersion was used for all the evaporations. The device shown in this section is similar to the device shown on section C of the main text (same process, cleaved at the same time) but differs by the fact that the GTI mirror was coated on a different evaporation run. The device operates at room-temperature emitting $> 100\text{ mW}$ of output power in CW operation (see Fig. S3a). No high-phase noise regime is observed for this device, as observed for the device described on the section C of the main text. The device shows single narrow RF beat-notes over the entire current range where multimode operation is observed (see Fig. S3a for the RF spectra and Fig. S3b for the optical spectra as a function of current at a fixed temperature), characteristic of comb regime operation.

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