Supplementary Information to "Frequency-modulated diode laser frequency combs at 2 μm wavelength"

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SUPPLEMENTARY NOTE I: SINGLE- AND MULTI-MODE OPERATION

As discussed earlier, several theoretical works have identified two prerequisites for self-starting FM comb generation in a semiconductor laser cavity: spatial hole burning (SHB) enabling multi-mode operation with dispersed modes, and a nonlinear phase synchronization mechanism provided by four-wave mixing (FWM) to ensure modal equidistance and low-phase-noise operation. While in most previously demonstrated FM combs the first condition of multi-mode lasing is easily achieved, here we observe a radically different behavior. Fig. S1 plots the normalized optical and intermode beat note frequency spectra in logarithmic scale as a function of injection current at room temperature acquired in two current ramp directions. In the first, the current was rapidly increased to 420 mA (>7 times threshold), and then slowly ramped down (Fig. S1a), whereas the second scan was in the opposite direction. Unexpectedly, the ramp-up experiment (Fig. S1b) yielded nearly single-mode emission even at currents exceeding threshold multiple times. The side-mode suppression ratio exceeds 15 dB for most cases, which is in stark contrast to QD/QDash, QCL or ICL devices promoting multi-mode operation shortly above threshold. A possible explanation of this QWDL behavior has been provided by Bardella et al.[1], and Dong et al.[2] Multi-mode lasing enabled by SHB is ascribed to fast sub-wavelength carrier gratings arising in the cavity due to standing-wave effects. These gratings destabilize single mode operation, and favor lasing on multiple lines. If the key requirement for the diffusion length to be short compared to the wavelength is not fulfilled, the spatial grating is smeared out and the laser tends to remain single-mode. Although for the QWDL it was predicted to lie in the micrometer range, the continuous presence of stimulated emission may significantly shorten the carrier lifetime[3]. Therefore, higher optical intensities in QWDLs operating at longer wavelengths are likely to enable SHB to occur and trigger multimode operation.

Experimental data seem to confirm these theoretical predictions. In the ramp-up scan, only at highest injection currents the optical spectrum becomes truly multimode with a multi-kHz wide and ≥50 dB strong electrical intermode beat note indicating the presence of FWM responsible for phase synchronization. At lower currents, a nearly-single mode exists, which rapidly switches its center wavelength due to a significant red-shift of the gain peak compared to the thermally-induced tuning of suppressed longitudinal modes. The ramp-down scan (beginning with an immediate increase to 420 mA) shows that once the laser enters the multimode regime, it stays in it over extended current ranges and allows for fine-tuning the spectrum for the most uniform envelope and narrowest electrical intermode beat note. This hysteretic comb-starting behavior was quite reproducible and low-noise regimes operating stably over hours were found at currents exceeding 340 mA once multi-mode operation was triggered. The origin of this behavior can be related to the SHB mechanism itself, and the assumption that the laser naturally tries to maximize its roundtrip gain[4]. Because the energy landscape of SHB is highly non-monotonic[5], different laser states can be accessed depending on the history of their development. The laser will simply settle into local maxima of the energy corresponding to different intensity profiles.

SUPPLEMENTARY NOTE II: REPETITION RATE LOCKING

In order to analyze the comb coherence properties using the phase-sensitive SWIFTs technique[6], it is useful to first stabilize the comb repetition rate f_{rep}. Light from the comb was optically detected by a fast 2 μm detector (Discovery Semiconductors, DSC-R202), and next it was amplified, mixed down to 20 MHz, and fed to a fast lock-in amplifier (Zurich Instruments, UHFLI). The built-in digital fast (300 kHz) phase locked loop (PLL) comprising a phase detector and PI controller was used to modulate the QWDL injection current and thus stabilize the repetition rate f_{rep}. When enabled, it allowed us to reach sub-hertz 3 dB linewidths, as shown in Fig. S2. Pronounced amounts of mechanical and electrical noise in the 60–300 Hz range can be eliminated through more elaborate environmental shielding of the device, whereas the servo bump at ∼300 kHz can be shifted by using a faster loop. In principle, electrical injection locking[7] can be used instead, however, microwave losses at ∼19 GHz become significant for non-rf-optimized structures. Therefore, it was more practical to use the PLL scheme.

SUPPLEMENTARY NOTE III: LASER GAIN AND DISPERSION

In the context of OFC generation, intracavity dispersion and gain flatness play a fundamental role. Particularly high

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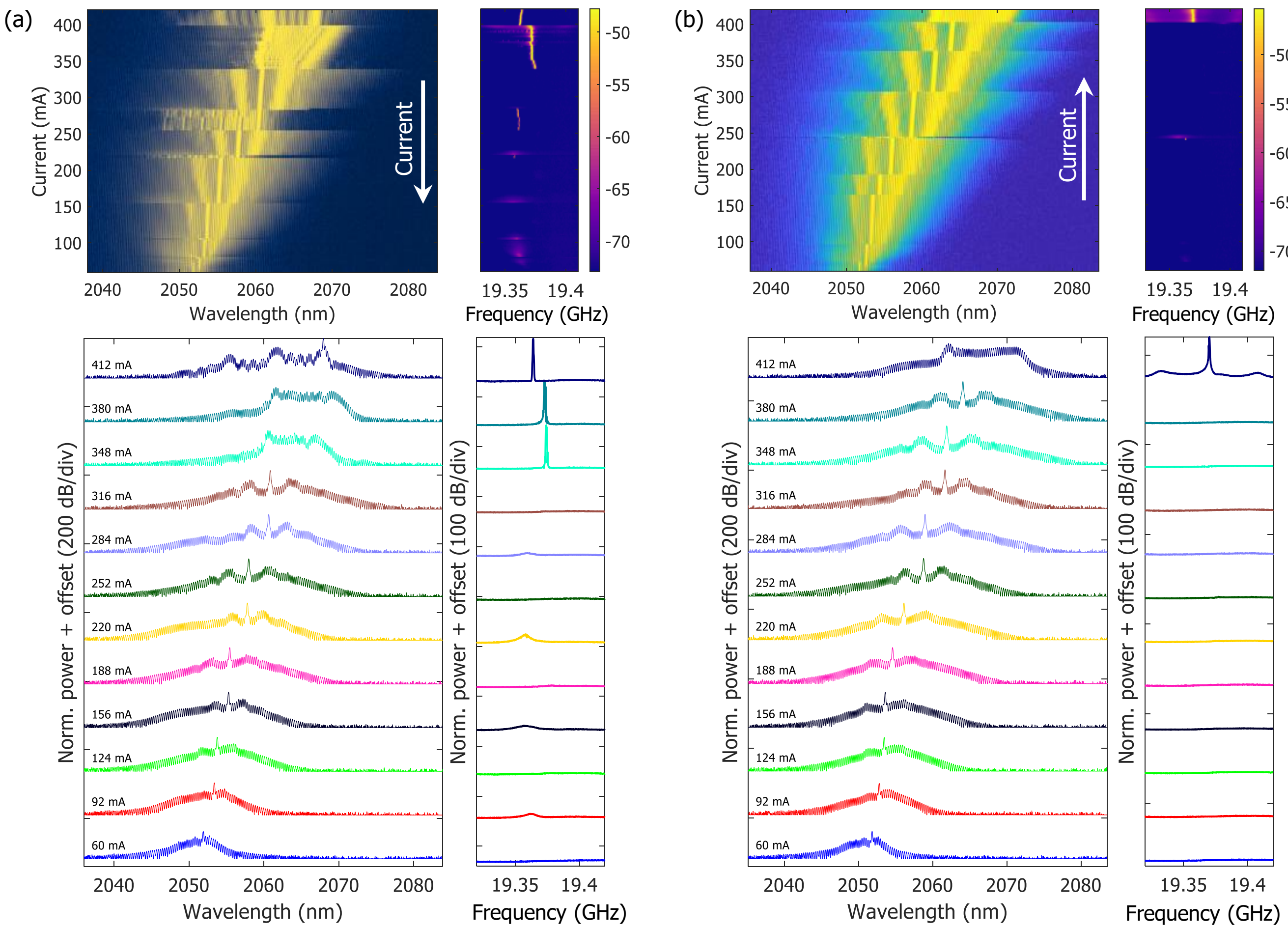

FIG. S1. Evolution of the optical and electrical intermode beat note spectrum when the injection current is tuned down (a) and up (b).

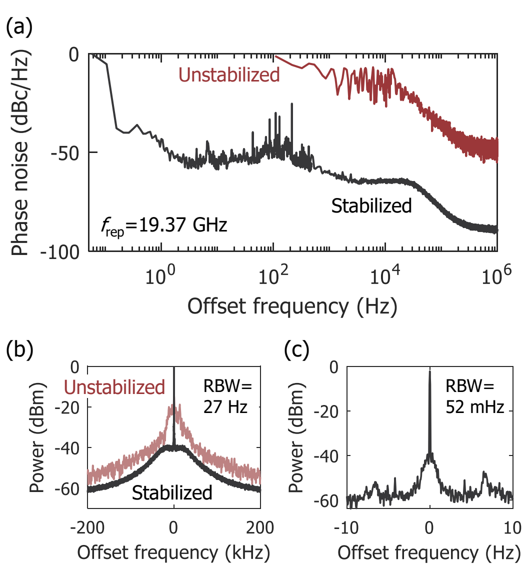

FIG. S2. Locking the QWDL comb repetition rate f_{rep} at 19.37 GHz. (a) SSB phase noise measurement of a free running device along with a stabilized using a fast optical phase locked loop (OPLL). (b) Frequency spectra of data in (a). Sub-Hertz linewidths are obtained.

GVD is unfavorable for comb formation, therefore attempts are made to engineer it through elaborate waveguide designs[8] and multi-layer dielectric coatings deposited on laser facets[9]. However, it would be interesting to evaluate these parameters before any optimization steps are taken. We employed the sub-threshold Fourier transform technique[10] by recording the emission spectrum in step-scan for the QWDL device biased below threshold ($\sim 0.95 I_{\mathrm{th}}$). Next, we compared the spectral amplitude and phase of the main and satellite interferogram bursts, as plotted in Fig. S3d. The calculated phase (Fig. S3a) accumulated over a 2 mm cavity roundtrip is convex and can be approximated by a second-order polynomial with a corresponding positive dispersion D_2 of $\sim$23300 fs^2, and a mean GVD of $\sim$5825 fs^2/mm. The accumulated dispersion is similar that reported for ICL cavities[11], yet many times higher than that reported for unoptimized QCL cavities[9]. It is unexpected that such relatively high dispersion values do not preclude stable comb formation.

The second derivative of the accumulated optical phase yields a spectrally-resolved cavity GDD (Fig. S3b), which in the lasing range shows relatively broad and locally flat regions of positive dispersion attributable to a smooth gain profile (Fig. S3c). The latter is slightly asymmetric, which may intro-

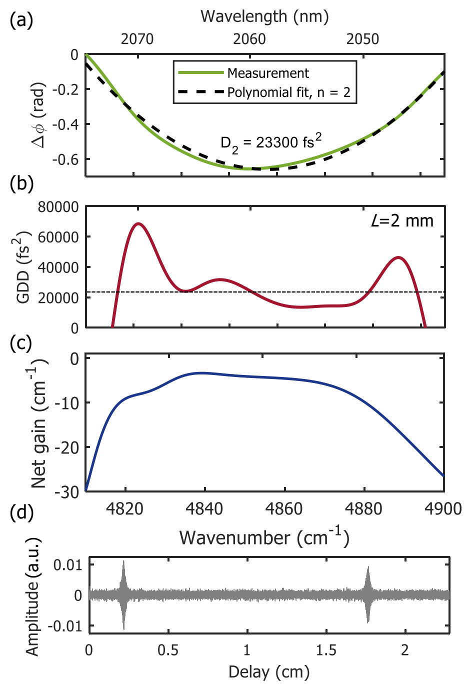

FIG. S3. Sub-threshold measurements of the 2 mm QWDL device. (a) Calculated spectral phase with a fitted positive second order dispersion profile. (b) Second derivative of the spectral phase in (a). Dashed line shows the fitted mean GDD. (c) Net modal gain. (d) Source data taken in step scan (300 ms of integration per step).

duce a Kerr nonlinearity contribution in phase locking above threshold, as postulated in a recent theoretical work[12].

SUPPLEMENTARY NOTE IV: TRANSMISSION SPECTRUM UNCERTAINTY

It would be interesting to see how the spectroscopic transmission uncertainty σ_T evaluated from rf amplitude fluctuations scales with relative dual-comb beat note power P (at a constant acquisition time, 1 ms for the reference and sample here). Note that the uncertainty depends on instabilities of the peak amplitudes and it cannot be improved by merely adding an rf amplifier. Instead, equalization of the spectral envelope and suppression of laser relative intensity noise is required.

Figure S4 plots the optical transmission uncertainty σ_T at 1 ms of integration. We find that the data points are governed by the well-known rule of precise DCS measurements[13]: to increase the precision (sensitivity) by a factor of 10, the beat note power must be increased 100 times (20 dB). The actual fit to the data suggests a factor of 150 (21.77 dB) for the required power, which can be attributed to many experimental factors. The main is probably imperfection of the digital phase correction. This figure, however, describes only the stability of the beat notes. Systematic errors introduced by the system drift, spectroscopic model inaccuracies, photodetector nonlinearities, and optical fringe noise may introduce errors in optical transmission greatly exceeding these predictions.

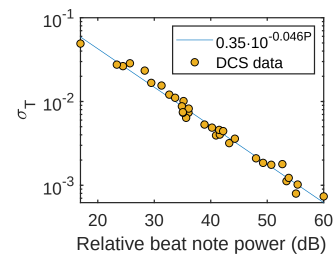

FIG. S4. Standard deviation of the optical transmission versus relative beat note power at 1 ms of integration. To increase the uncertainty from the percentage to per-mille regime, the beat note's power must be increased by 21.77 dB ($\sim$150 times).

SUPPLEMENTARY NOTE V: CURRENT AND THERMAL TUNING

The spectroscopic coverage of the comb can be increased by exploiting injection current and thermal tuning. Both allow to perform measurements beyond the sparse comb tooth spacing, which is particularly important for detecting narrow-band gas species such as carbon dioxide. While injection current tuning allows for smaller wavelength shifts compared to thermal, it occurs on a much faster time scale. Figure S5 plots a zoomed view of Fig. S1a in the current region where the comb becomes broadband, relatively flat and broadly tunable (344 – 397 mA). As evident from the figure, wavelength scans close to a full f_{rep} are possible, however, at higher injection currents the intermode beat note starts to become slightly noisier and broader. Future dispersion engineering of the laser cavity should help to promote low phase noise operation over larger current ranges. The current tuning coefficient estimated from the spectral map is $\Delta\lambda/\Delta I$ = 6.6 pm/mA.

Another possibility to shift the comb's center wavelength is thermal tuning (Fig. S6). Although the obtainable wavelength shifts are comparable to the comb bandwidth, step-changing the temperatre setpoint may require even a minute or more for the heatsink temperature to fully stabilize. Here, we cooled the device's submount from 20°C to 15°C. At nearly the same injection currents ($\sim$ 360 mA) and narrow near-kilohertz intermode beat note linewidth, the spectrum was shifted with very little changes of the mode structure by 15.6 cm^{-1} (6.62 nm / 468 GHz). This tuning mechanism is determined predominantly by the temperature dependence of the maximum gain wavelength $\Delta\lambda_{g_{max}}/\Delta T$. Here, the comb center wavelength tunes by $\Delta\lambda_c/\Delta T \approx$ 1.2 nm/K, which is in good agreement with the value reported previously for Fabry-Pérot devices made from the same materials (1.39 nm/K)[14].

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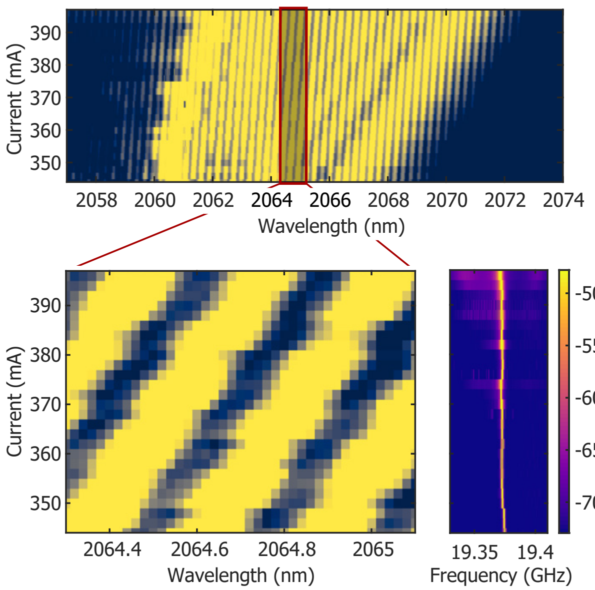

FIG. S5. Injection current tuning in the broadband and quasi-flat frequency comb regime. The bottom left panel shows a zoom on the central part of the optical spectrum revealing the feasibility of a gapless scan. Unfortunately, at highest injection currents, the laser enters a slightly phase-noisy regime visible as a pronounced noise pedestal of the microwave intermode beat note (bottom right panel, dB scale).

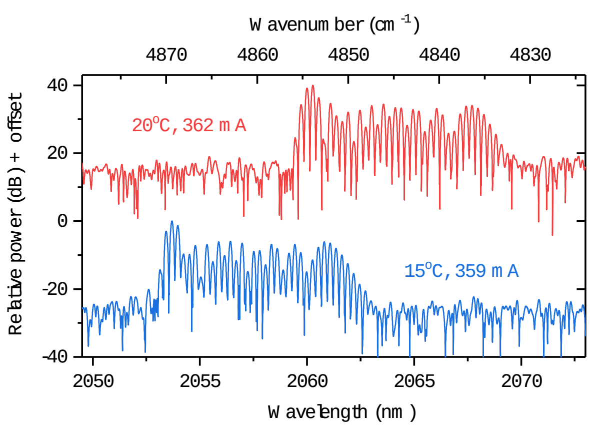

FIG. S6. Thermal tuning of the laser spectrum in a low-noise frequency comb regime measured with an FTIR.

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