Mid-infrared frequency comb from a ring quantum cascade laser: supplementary material

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\textbf{1. MAXWELL-BLOCH MODEL}

Let us consider a Maxwell-Bloch model with the modal decomposition of the cavity field, as used recently to model QCL-based frequency combs [1,2].

The difference between a Fabry-Perot and a ring cavity is especially clear when considering a Maxwell-Bloch model with the modal decomposition of the cavity field, as used recently to model QCL-based frequency combs [1,2]. In the model, the fundamental equation describing the amplitude of the \( n \)-th mode in the comb is expressed as:

\[
\dot{A}_n = \{G_n - 1 + iD_n\}A_n - G_n \sum_{klm} C_{kl} B_{kl} A_k A_l A_m \kappa_{n,k,l,m} \tag{S1}
\]

where \( A_n \), \( G_n \), and \( D_n \) correspond to the amplitude, the gain and the dispersion of the \( n \)-th mode in the cavity, respectively. The coefficient \( B_{kl} \) describes the effective bandwidth of the four wave mixing, \( C_{kl} \) defines the complex amplitude of population oscillation, and \( \kappa_{n,k,l,m} \) is the intermodal overlap in real space.

In the ring cavity, the envelope of mode \( n \) traveling in the \( z \) direction is expressed as \( \exp(-ik_nz) \), where \( k_n \) is the wavevector. Thus, \( \kappa_{n,k,l,m} \) in the ring cavity is defined as:

\[
\kappa_{n,k,l,m} = \frac{1}{L} \int_0^L \exp(-ik_nz) \exp(-ik_kz) \exp(ik_lz) \exp(ik_mz)dz \tag{S2}
\]

where \( k_n \), \( k_k \), and \( k_l \) correspond to the wavevectors of modes \( n \), \( k \), and \( l \), respectively. Under the rotating-wave approximation, the \( \kappa_{n,k,l,m} \) equals to 1 for all the coupling pairs. Compared with the FP cavity, the ratio between the self-saturation and the cross-saturation is lower (the ratio equals to 1.5 and 1 for the FP and the ring cavities, respectively), thus leading to a stronger competition between the single mode and the multimode operations. However, the ring cavity shows a much increased non-degenerate four wave mixing to self-saturation ratio (1 and 1/3 for the ring and FP cavities, respectively), which is expected to enhance the stability of the frequency comb operation in the ring cavity.

\textbf{2. DETAILS OF THE DISPERSION MEASUREMENT}

The group velocity dispersion (GVD) was obtained using the Fourier transform method [3]. In our case, due to the difficulty in extracting the light output from the ring waveguide structure, instead of using the ring, we used a Fabry–Pérot (FP) cavity device. However, both structures should exhibit similar GVD for the same waveguide width according to the numerical simulation. In this method, the GVD is calculated based on the Fourier transform of the subthreshold emission spectrum from the device. For the measurement, we used a Fourier transform spectrometer with a highest resolution of 0.075 cm\(^{-1}\) and a liquid nitrogen cooled photoconductive MCT detector. The schematic of the experiment setup is shown in Fig. S1 (a) and the first sidelobe of the interferogram used for the dispersion calculation is shown in Fig. 1 (b).

Fig. S1. (a) Schematic of the experiment setup for the GVD measurement. (b) First sidelobe of the interferogram from the FP
3. NUMERICAL SIMULATION OF THE OUTPUT COUPLING EFFICIENCY FROM THE RING WAVEGUIDE

To calculate the output coupling efficiency from the ring waveguide, we performed a 2D numerical simulation of the whole waveguide structure using the RF module of COMSOL Multiphysics (Fig. S2 shows the coupled region of the structure and the mode distribution for the wavenumber of 1337 cm$^{-1}$ that is the center wavenumber of the lasing spectrum). In the simulation, the mesa was enclosed by the air regions with scattering boundary condition. The radius of the ring is 600 $\mu$m, the width is 8 $\mu$m, and the gap between the ring and air is 10 $\mu$m which is close the experiment value. The corresponding output coupling efficiency is expresses as

$$\eta = \frac{\iint E_z^2(x, y) \, dx \, dy}{\iint E_{all}^2(x, y) \, dx \, dy} \quad (S3)$$

where $E_z$ is the calculated steady state transverse electric field distribution. Due to the close proximity, part of the light tunnels into the air region as shown in Fig. S2, contributing to the output efficiency.

Fig. S2. Calculated transverse electric field $E_z$ distribution at a wavenumber of 1337 cm$^{-1}$.

4. DETAILS OF THE DEVICE STRUCTURE

The uniformity of the ring width and and the Fe:doped InP regrowth quality were characterized by the scanning electron microscopy (SEM). The cross-sectional SEM images of two orthogonal directions (labelled in Fig. S3 (a)) are shown in Fig. S3 (b) and Fig. S3 (c), corresponding to positions A and B, respectively. Both directions show a ridge width of ~ 8 $\mu$m, demonstrating good uniformity across the whole ring waveguide structure. Meanwhile, both directions show a nice regrown Fe:doped InP enclosing the waveguide.

Fig. S3. (a) Sketch of the device structure with the cleaving directions indicated by the orthogonal dashed red lines. (b) Cross-sectional SEM image of the ring waveguide at position A. (c) Cross-sectional SEM image of the ring waveguide at position B.

5. Optical spectra for the clockwise travelling mode

As mentioned in the main text, the device was systematically studied while driven into the CCW mode. We found the spectral characteristic in CW mode to be single mode, as displayed in the figure S4. Switching between comb and single mode operation was also observed in ring QCL in ref. [4]. We speculate that the lack of symmetry between CC and CCW operation arises from residual feedback from the facet.

Fig. S4. Optical spectra for the clockwise travelling mode at different currents. Single mode emission is observed in the whole current dynamic range.

6. TEMPERATURE DEPENDENCE OF THE RING COMB BEATNOTE

Fig. S5. Intermode beatnote spectra for the counterclockwise mode as a function of temperature.

7. PHASE MEASUREMENT OF REFERENCE LASER BY SWIFTS

Broadband frequency combs based on mid-infrared QCLs exhibit a predominantly parabolic phase, with the quadratic coefficient scaling inversely proportionally to the locked bandwidth. Ripples on the curve prevent us however from directly assuming a parabola; the recovered phase over the narrow interrogation bandwidth, and hence the reconstructed temporal behavior, will depend strongly on the local structure.

We hence pre-characterised the reference laser using SWIFTS (shifted wave interference Fourier transform spectroscopy), details on which can be found in [5,6], which measures the phase differences between neighbouring through a coherent demodulation of the spectrally resolved beat signal on a fast detector. Owing to a slow switching type instability, occurring on
millisecond to second timescales, our measurement suffered from spiking behavior and a hence greatly elevated noise floor.

Nonetheless, as the lasing behavior does not deviate significantly between the states, we are able estimate statistically the average spectral characteristic and apply bounds over the range of interest. We record 38 SWIFTS traces sequentially, with a total acquisition time on the order of 30 minutes. From these, we use Monte-Carlo/bootstrapping (full sample population resampling with replacement) to obtain a median expectation and 5-95% confidence intervals on the measured phase differences, calculated for each pair of modes. Statistical independence is assumed between mode pairs and between measurements.

The phase curve is obtained by a summation of the phase differences, centered at the arbitrarily defined center frequency of the ring laser, and the confidence intervals propagated along. Fig.S6 shows the result. Note that the central phase has a zero value and a zero uncertainty by definition. The uncertainty then increases as you move further from that tone as the error in the estimate accumulates. These are the uncertainties shown in Fig. 5 of the main paper.

For reference, we have plotted in (b) the uncertainty as a function of wavenumber over the ring laser’s optical bandwidth. It can be seen that for the most 5 most important contributing modes, the uncertainty level is at most 0.11 \( \pi \) radians. Nonetheless, the phase of the reference laser is by far the dominant source of uncertainty in our measurement.

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\[ \omega'_c = \omega'_{c,0} + n \omega' \]

\[ \omega''_c = \omega''_{c,0} + m \omega'' + m(2 \omega'' + \Delta \omega) \]  

Multiheterodyne beats will be generated at:

\[ \omega_{\text{mult}} = \omega_{c,0} + \omega' (n - 2m) - m\Delta \omega \]

Depending on the offset frequencies, indices:

\[ |\Delta \omega_{c,0}| \leq B, n = 2m \]  

\[ |\Delta \omega_{c,0}| \geq \omega', B, n \pm 1 = 2m \]

Where \( B \) is the electronic detection bandwidth. The signal will be:

\[ V \propto \sum_{n,c} B^c_{n} e^{i \omega_{c,0} t} \exp \left( i \phi^{c}_{n} - \phi^{c}_{n} + \phi_{c,0} \right) + c.c. \]  

Details on other implementations can be found for example under [7,8]. After the coherent correction, which pins both \( \Delta \omega_{c,0} \) and \( \Delta \omega \) in the case both lasers are frequency combs, the Fourier terms composing the multiheterodyne are found by directly evaluating:

\[ c_{s} = \sum_{j} V_{j} \exp \left( -i \omega_{\text{mult},a} t_{j} \right) \]

where the time vector \( t \) must be tracked through the entire calculation and \( j \) is the time index. The phase difference term \( \phi_{\text{ring}} - \phi_{\text{ref}} \) will carry a positive sign if \( f_{\text{ring}}' < f_{\text{ref}}' \) and negative if \( f_{\text{ring}}' > f_{\text{ref}}' \).

An example of a typical acquisition is plotted in Fig. S7 (a), with a comparison of the raw and corrected spectra shown in (b). Though interspersed with jumps in the frequency of the reference laser, the more prevalent periods of stability remain useful for our analysis, as evidenced by the striking effectiveness of the two-parameter correction.

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The trajectory of the coefficients are plotted in polar coordinates in Fig 8. (a) for the same slice. Crucially, none rotates about the origin, demonstrating the equidistance between the multiheterodyne tones, and hence the equidistance between the two combs themselves. As a corollary, the phase differences between the multiheterodyne tones is fixed.

In the opposite case, a non-equidistance greater than the resolution bandwidth would manifest itself both in the form of a rotation about the origin, and of a zero-bound trajectory if instead considered in the form of a cumulative average. This is shown certainly not to be the case, by the facts that a) the overall average value, calculated with all samples, sits in the centre of each point cloud, and b) the signal to noise ratio of the of the multiheterodyne spectra is shown to increase at a constant rate. Equivalently, the Allan deviation in Fig S8 b) diminishes at a rate of $1/\tau$ corresponding to white frequency noise, with no sign of inflection point at our timescales. This justifies the averaging.

One could also consider a deviation in equidistance below the resolution bandwidth of the acquisition, which would show only a slow drift in the phase and amplitude. In this case, however, the different measurements, in our case taken several seconds apart, would not be expected to agree.

References