Mode-locked ultrashort pulses from an 8 µm wavelength semiconductor laser

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Quantum cascade lasers (QCL) have revolutionized the generation of mid-infrared light. Yet, the ultrafast carrier transport in mid-infrared QCLs has so far constituted a seemingly insurmountable obstacle for the formation of ultrashort light pulses. Here, we demonstrate that careful quantum design of the gain medium and control over the intermode beat synchronization enable transform-limited picosecond pulses from QCL frequency combs. Both an interferometric radio-frequency technique and second-order autocorrelation shed light on the pulse dynamics and confirm that mode-locked operation is achieved from threshold rollover current. Being electrically pumped and compact, mode-locked QCLs pave the way towards monolithically integrated non-linear photonics in the molecular fingerprint region beyond 6 µm wavelength.

The discovery of ultrashort light pulses has led to numerous breakthroughs in science and technology, including frequency combs\textsuperscript{4,5} and refractive surgery in ophthalmology\textsuperscript{6}. Nowadays, optical pulses are routinely generated in mode-locked lasers operating in the visible or near-infrared range\textsuperscript{7,8}. Currently, large efforts are aimed at bringing ultrafast laser science in the mid-infrared (MIR) region to a similarly high degree of maturity\textsuperscript{9,10}. Due to the lack of suitable gain media, methods for the generation of pulses in the molecular fingerprint region beyond 5 µm wavelength have so far relied on non-linear downconversion of near-infrared pulses\textsuperscript{11}. Established techniques such as optical parametric oscillators\textsuperscript{12} or difference frequency generation\textsuperscript{9,10} either require sophisticated optical setups with tabletop dimensions or are restricted to mW-level of output power.

Quantum cascade lasers\textsuperscript{11} (QCL) have matured to become the dominant MIR laser source. While being microchip-sized and electrically pumped, they are capable of producing Watt-level average power\textsuperscript{12,13}. Quantum engineering of the active region allows to tailor the emission wavelength throughout the entire mid-infrared region. Hence, harnessing high-performance QCL technology for the generation of MIR pulses represents a long-sought milestone in ultrafast laser science. Mode-locked QCLs could serve as monolithic pump lasers for microresonators and resonant supercontinuum generation\textsuperscript{14} paving the way towards broadband and high-brightness frequency combs. So far, the sub-picosecond carrier transport in QCL active regions has constituted a seemingly insurmountable obstacle for the formation of short light pulses\textsuperscript{10-14}. To date, the only successful attempt of mode-locking in monolithic MIR QCLs was observed using a specially designed active region with strongly enhanced upper-state lifetime of the lasing transition\textsuperscript{15}. However, the necessary design modifications limited mode-locked operation to cryogenic temperatures and peak power below 10 mW, thus impeding their practical use.

In this work, we demonstrate the generation of transform limited picosecond pulses in high-performance 8 µm wavelength QCLs at room temperature both experimentally and theoretically. Mode-locking is achieved by electrically modulating the intracavity loss using a short modulation section designed for efficient radio-frequency (RF) injection (Fig. 1a). In order to achieve the large modulation depth required for stable mode-locking, close attention has to be paid to the band structure of the QCL active region. This effect is illustrated in Figs. 1a,b. As the bias applied to a standard QCL structure is decreased, it does not switch to absorption at the lasing wavelength, but becomes nearly transparent for the intracavity light due to a bias-dependent shift of the electronic levels, known as Stark effect (Fig. 1b). Hence, the modulation depth is severely limited in standard QCL designs. For this reason, we employ a bi-functional active region whose lasing wavelength and absorption wavelength at zero bias were matched to each other\textsuperscript{16} (Fig. 1c). This strategy allows to overcome the aforementioned limitations of the modulation depth caused by the Stark shift. Most importantly, the bi-functional design shows excellent overall performance, which is competitive with other state-of-the-art designs. A 3.5 mm long device mounted epitaxial-side up emits more than 130 mW average power in continuous wave at room temperature (Fig. 2a).

As a first step towards pulse generation, it is essential to determine the optimal modulation frequency. For this purpose, mode-locking can be seen as synchronization of coupled oscillators\textsuperscript{17} (Fig. 1d). Each pair of neighboring cavity modes creates a beating at their difference frequency, which is equal to the cavity roundtrip frequency.
These beatings can be seen as oscillators coupled by the non-linearity of the gain medium. Thanks to this coupling, the cavity modes of a free-running QCL can be locked together without modulation, thus giving rise to a self-starting frequency comb. Yet, this kind of frequency comb does not emit isolated pulses, but rather a quasicontinuous wave accompanied by a strong linear frequency chirp. This corresponds to anti-phase synchronization and will be called frequency modulated (FM) comb in the following. In contrast, in-phase synchronization of the in-phase and anti-phase states synchronize at different frequencies depending on the coupling. As a consequence, while the cavity roundtrip frequency of the FM QCL comb \( f_{\text{rep}}^0 \), may seem like a reasonable choice, we expect the optimal modulation frequency for generating pulses to differ from \( f_{\text{rep}}^0 \).

In order to investigate these two synchronization states experimentally, we start by operating the laser well above its threshold current. Subsequently, the DC bias of the modulation section is decreased to 2.8 V, where the large absorption caused by the bifunctional design (Fig. 1c) brings the QCL just slightly below lasing threshold. In these conditions, modulation at the right frequencies can provide enough additional gain to reach threshold. Fig. 1f shows the laser power depending on modulation frequency and power. At 33 dBm modulation power, the QCL reaches threshold when modulating close to \( f_{\text{rep}}^0 \). Strikingly, a second modulation frequency where lasing occurs is observed almost 60 MHz higher than \( f_{\text{rep}}^0 \), as predicted by the picture of synchronized oscillators. Both the range around the two synchronization frequencies, where lasing is observed, as well as the optical power grow upon increasing the modulation power.

Even more insight is provided by using a two-photon quantum well infrared photodetector (2-QWIP) to detect the emitted light. The signal of the 2-QWIP is proportional to the square of the intensity. This allows to identify which modulation frequency leads to in-phase and which to anti-phase synchronization (Fig. 1g). Again, two lobes
Figure 2 – Mode-locked pulses from an 8 µm wavelength QCL. a: SWIFTS characterization of the QCL operated close to lasing threshold. The laser is modulated at the in-phase synchronization frequency and at 37 dBm power level. b: Reconstructed time-domain signal of the QCL, showing a train of transform limited pulses with 6.5 ps FWHM. c: Simulation of the QCL using the coherent Master equation described in supp. section 1. d: Interferometric autocorrelation (IAC) of the QCL pulses close to threshold. Red dots: envelope of the IAC reconstructed using SWIFTS. e: IAC at the rollover current, still displaying the 8:1 ratio. The second burst at a delay equal to the cavity roundtrip time is due to the interference of subsequently emitted pulses. Its peak value of 8 provides another proof for the coherence of the pulses because phase-decoherence would smear out the fringes of the IAC and thus decrease the peak value to smaller than 8. Inset: zoom on the interferometric fringes.

appear around \( f_{\text{exp}} \) and 60 MHz above. Yet, the 2-QWIP signal is more than an order of magnitude larger in the lobe at higher \( f_{\text{mod}} \). At this frequency, the laser operates in the in-phase synchronization regime and emits intense pulses, which leads to a strongly increased 2-QWIP signal.

In order to unequivocally prove mode-locking, we employ two independent methods to characterize the pulse dynamics at three points of operation from threshold up to the rollover current. Firstly, an interferometric RF technique called ‘Shifted wave interference Fourier transform spectroscopy’ (SWIFTS) is used to measure the phases of the QCL spectrum (details in Methods section). This information not only enables the reconstruction of the temporal waveform, but also allows to assess the phase-coherence of the pulses and whether they form a frequency comb. Secondly, we measure the interferometric autocorrelation (IAC) of the pulses using the 2-QWIP, which constitutes an additional well-established proof for mode-locking and the pulse width. Fig. 2a shows the SWIFTS characterization of the QCL operated close to threshold. In contrast to the free-running laser, the intensity spectrum consists of a single Gaussian-shaped lobe. The SWIFTS spectrum represents the part of the intensity spectrum which is beating exactly at the modulation frequency. Since the SWIFTS spectrum has the same shape as the intensity spectrum over its entire span, the QCL generates a frequency comb whose repetition frequency is given by the modulation frequency. The intermodal difference phases \( \phi \), which correspond to the spectral group delay, are synchronized almost perfectly in-phase. Hence, all parts of the spectrum have the same group delay and form a pulse. Indeed, the reconstructed waveform in Fig. 2b shows the emission of 6.5 ps short pulses. The full-width-half-maximum (FWHM) of the reconstructed pulses is given by the transform limit of the spectrum in Fig. 2a, indicating that there is negligible chirp in the pulses. In these conditions, the peak power reaches almost 250 mW, which constitutes an enhancement of more than 12 compared to the average power of 20 mW. In order to model the cavity dynamics, we use a fully coherent master equation (supp. section 1). This single equation for the complex field replaces the entire Maxwell-Bloch system and reliably predicts the spectral shape, phase relationship and pulse width observed experimentally (Fig. 2a). Furthermore, it allows experimentally unavailable analyses, e.g. the influence of dispersion and nonlinearities (supp. Figs. 2,3,7).
The IAC close to threshold (Fig. 2f) shows a prominent peak at zero path difference caused by constructive interference of the pulses after the Michelson interferometer. The ratio of this peak to the background at a delay larger than the pulse width is 8:1, which is generally regarded as the smoking gun of mode-locked pulses. Encouragingly, the measured IAC is in excellent agreement with the expected IAC, which was calculated using the pulses obtained by SWIFTS (red dots in Fig. 2f). This confirms successful mode-locking and the retrieved pulse width.

The generation of pulses becomes increasingly challenging at higher gain current. Due to gain saturation, the wings of a pulse experience more gain than the peak. This effect leads to pulse broadening and can destabilize mode-locking. Fortunately, the large modulation depth provided by the bi-functional quantum design enables mode-locking over the entire lasing range from threshold to rollover. The IAC traces at 3.7 kA/cm² and at the rollover current still show the required peak-to-background ratio of 8:1. The pulses at rollover are slightly broadened to roughly 12 ps, which is attributed partially to a slight chirp and partially to the gain saturation effect mentioned above (supp. Fig. 6). Yet, the average power is greatly increased to 62 mW, which results in over 430 mW peak power and 5 pJ pulse energy - more than an order of magnitude higher than recent reports of comparable mid-infrared semiconductor lasers emitting at shorter wavelengths.

Another fascinating aspect of bi-functional quantum design is the possibility to monolithically integrate ultrafast photodetectors. While this is particularly important in applications such as photonic integrated circuits, it also provides a tool to measure the beatnote with very large signal-to-noise ratio directly on the chip (Fig. 3a). This provides crucial information about the type of synchronization state and about its stability. Fig. 3b shows the first three harmonics of the beatnote in the free-running and the actively mode-locked regime. In the latter conditions, the beatnote amplitudes increase by 19 dB due to the much larger amplitude modulation. The zoom on the first harmonic beatnote (Fig. 3c) allows to assess the phase-coherence and stability of the frequency comb. The free-running QCL is operating in the anti-phase state showing a weak beatnote at $f_{\text{rep}}$. Previous work has shown that a weak electrical modulation can be used to lock and stabilize the beatnote of the anti-phase state. However, the situation is very different when applying strong modulation at $f_{\text{rep}}$. In this case, the modulation enforces an AM waveform, which is contrary to the natural anti-phase behaviour of the laser. As a result, the beatnote of this waveform is not phase-locked, as indicated by the pedestal around $f_{\text{mod}}$. The situation changes completely, when the modulation frequency is tuned to the synchronization frequency of the in-phase state ($f_{\text{rep}}+33$ MHz). There, the strong modulation is in consensus with the natural behavior of the laser, leading to a phase-locked frequency comb with narrow beatnote. This can also be seen in Fig. 3d, which shows the laser beatnote while tuning the modulation frequency across the in-phase and anti-phase synchronization frequencies. While the frequency of the beatnote is controlled by the modulation over the entire span, a phase-locked comb is only generated around the in-phase synchronization frequency.

In conclusion, our experiments provide unambiguous proof for the generation of mode-locked pulses in high-performance QCLs at room temperature - a goal which remained elusive since the invention of the QCL - and confirm stunning similarities to synchronization in coupled oscillators. These mode-locked QCLs constitute the
first compact and electrically pumped source for ultrashort pulses beyond 5 µm wavelength, demonstrating that they are a highly promising technology for ultrafast laser science in the long-wave infrared region. The availability of such a source paves the way towards a semiconductor-based platform for non-linear photonics, potentially enabling broadband mid-infrared frequency combs and supercontinuum generation.

Methods

QCLs optimized for RF modulation: The QCLs are processed as buried heterostructures. The width of the QCL ridges is 12 µm and the facets are left as cleaved. The area of the top contact of the modulation section is minimized to decrease its parasitic capacitance, which increases the RF injection efficiency. Ground contacts for the modulation are provided by etching through the Fe-doped InP layer next to the laser ridges. The modulation signal is provided by a HP8341B synthesized sweeper, amplified up to roughly 5 W and injected via coplanar tips. The insertion loss at 12 GHz is 14 dB including cables and bias-tee.

SWIFTS and IAC: The light emitted by the QCL is shone through a Bruker Vertex 70v FTIR spectrometer. In order to perform SWIFTS, the light is then detected by a home-built fast QWIP at the exit of the FTIR spectrometer. In order to perform SWIFTS, the pulses at the exit of the FTIR using the 2-QWIP and recording trigger of the FTIR are used to record the SWIFTS and intensity in-oscillator. A Zurich Instruments HF2LI lock-in amplifier and the subsequently amplified and mixed down to roughly 10 MHz using a local oscillator.

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Author contributions

J.H. processed the QCLs and carried out the experiments. B.S. and J.H. built up the SWIFTS and IAC setups. N.O. and B.S. developed the simulation tool. M.P. carried out the temporal reconstruction using the IAC data. H.S. provided the 2-QWIP. J.H. wrote the manuscript with editorial input from N.O., B.S., G.S and F.C. All authors analysed the results and commented on the paper.