High performance quantum cascade laser frequency combs at $\lambda \sim 6 \mu m$ based on plasmon-enhanced dispersion compensation

**SARGIS HAKOBYAN,**$^{1,*}$ **RICHARD MAULINI,**$^{1}$ **STÉPHANE BLASER,**$^{1}$ **TOBIAS GRESCH,**$^{1}$ **ANTOINE MULLER,**$^{1}$

$^1$Alpes Lasers SA, Avenue des Pâquiers 1, CH-2072 Saint-Blaise, Switzerland
* sargis.hakobyan@alpeslasers.ch
http://www.alpeslasers.ch/

Abstract: We demonstrate Quantum Cascade Laser (QCL) optical frequency combs emitting at $\lambda \sim 6 \mu m$. A 5.5 $\mu$m-wide, 4.5 mm-long laser exhibits comb operation from -20 °C up to 50 °C. A maximum output power of 300 mW is achieved at 50 °C showing a robustness of the system. The laser output spectrum is $\sim 80 \text{ cm}^{-1}$ wide at the maximum current, with a mode spacing of 0.334 cm$^{-1}$, resulting in a total of 240 modes with an average power of 0.8 mW per mode. To achieve frequency comb operation, a plasmonic-waveguide approach is utilized. A thin, highly-doped Indium Phosphide (InP) layer is inserted in the top cladding design to compensate the positive dispersion of the system (material and waveguide). This approach can be further exploited to design QCL combs at even shorter wavelengths, down to 4 $\mu m$. Different ridge widths between 2.8 and 5.5 $\mu m$ have been fabricated and characterized. All of the devices exhibit frequency comb operation. These observations demonstrate that the plasmonic-waveguide is a robust and reliable method for dispersion compensation of a semiconductor laser systems to achieve frequency comb operation.

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1. Introduction

Frequency combs have become a cornerstone of the spectroscopy and metrology communities, due to their unique properties of coherently linking the microwave and optical frequencies, by providing low-noise equidistant comb teeth at known frequencies [1, 2]. They are being used as frequency rulers for precise measurement of various optical and radio signals [3], in atomic clocks [4, 5], spectroscopy [6–8], and many more applications. High repetition rate frequency combs are of particular interest for applications such as low-noise microwave signal generation [9, 10], calibration of astronomical spectrographs [11, 12] etc. Frequency combs in the Near Infrared (NIR) optical region are profoundly developed, many technologies are being used for the latter, such as fiber laser systems [13], solid-state lasers [14], optically pumped semiconductor lasers [15, 16], microring resonators [17, 18] etc. However, achieving high repetition rate frequency comb operation has been a challenge for most of the technologies mentioned above [19] (except microring resonators). Yet another challenge, and maybe even more profound one, is the limitation to the NIR optical spectra, while most of the interesting molecules have absorption spectra in the Mid-Infrared (MIR) spectral region. Based on nonlinear optics several approaches are reported to achieve frequency comb operation in the MIR [20, 21]. However, all of the mentioned approaches have the drawback of non-robustness, complexity of the system. Quantum Cascade Laser (QCL) based frequency combs seem to be the ideal solution for both limitations [22]. Flexible bangap engineering allows one to design and make lasers with almost any specific emission spectrum in the MIR region (from 4 $\mu m$ to $> 10 \mu m$), as well as in the THz region where several comb applications have been reported [23, 24]. The intrinsic size of the waveguide chip automatically leads to multi-GHz repetition rates. The major challenge to
generate optical frequency combs with QCLs is the limitation due to the material and waveguide dispersions [22], especially at short wavelengths, closer to the bandgaps of the constituent materials. Various methods have been developed for dispersion compensation of MIR QCLs for frequency comb operation including integrated Gires-Tournois interferometer (GTI) mirror [25], coupled waveguides [26], and dispersion-compensating facet coatings [27,28]. However, the main drawbacks remain either the compactness and integrity of the method, or complexity of engineering and fabrication. Recently, plasmonic-waveguide approach has been demonstrated as a successful method for dispersion compensation and frequency comb operation of QCLs at 7.8 \( \mu \text{m} \) [29]. However, for shorter wavelengths, this approach appears to be challenging due to the associated waveguide losses that will be discussed in this paper.

Here we demonstrate QCL frequency comb centered at 6 \( \mu \text{m} \) that is utilizing a novel type plasmonic-waveguide approach to compensate the dispersion. 6 \( \mu \text{m} \) is a very interesting spectral range for detailed analysis of dynamical changes of proteins [30,31], it corresponds to the amide I vibration band of proteins and alloys. Detailed experimental analysis and theoretical simulation analysis has been performed to explain the behavior and to achieve the best structure for dispersion compensation, as well as keeping the waveguide losses at a moderate level. The emission spectrum is \( \sim 80 \text{ cm}^{-1} \) wide at the maximum current, covering the wavelength range from 5.87 \( \mu \text{m} \) to 6.15 \( \mu \text{m} \), with a mode spacing of 0.334 cm\(^{-1}\), resulting in a total of 240 modes with an average power of 0.8 mW per mode. Different ridge widths have been processed, ranging from 2.8 \( \mu \text{m} \) to 5.5 \( \mu \text{m} \). In all the cases frequency comb operation was observed up the maximum temperature of 50 °C indicating the robustness of the method for dispersion engineering. This is a great advancement towards achieving stable, reproducible QCL combs at 6 \( \mu \text{m} \) optical range.

2. Waveguide simulations

To understand the dispersion profile of the waveguide, multiple parameters of the QCL waveguide growth have been varied and simulated. In order to reduce the waveguide dispersion for 5.8 to 6 \( \mu \text{m} \), we introduce a thin layer of highly doped InP between the upper cladding layers. Due to the coupling between the waveguide mode centered on the active region and the plasmon mode appearing in that doped layer, the dispersion is expected to be strongly affected. For simulating the electric field of the lasing mode in the waveguide, the finite element method is used. The simulated waveguide facet is presented in Fig. 2. In the same plot, the parameters that are being modified are specified with black and white arrows.

First, we designed a plasmon-waveguide for reducing the dispersion of a waveguide with ridge width of 5.5 \( \mu \text{m} \), the resulting Group Velocity Dispersion (GVD) from the simulation for no plasmon-layer (blue) and with plasmon layer (red) structure are plotted in the Fig. 1 left. For all the waveguide simulations, the calculated dispersion takes into account material and waveguide contributions, the gain contribution is not considered in the simulations.
The grey areas in Fig. 1 highlight the range of the laser’s measured emission spectrum. The mean dispersion is reducing by about $\sim$500 fs$^2$/mm from 800 to 300 fs$^2$/mm. However, due to the interaction with the plasmonic layer, the waveguide losses are increased by 0.55 cm$^{-1}$ (Fig. 1 right). The norm of the electric field distribution in the waveguide for various wavelengths is shown in Fig. 2.

In this figure, cl4 is the highly doped InP layer that serves as a plasmonic-waveguide, cl5 is
low doped InP layer that serves as a spacer between the cl4 (plasmonic layer) and the AR (to control the interaction of the mode with the plasmon). The field penetration to plasmonic-layer is higher with increasing wavelengths (compare Fig. 2a - c).

With Fig. 1, it becomes evident that not only the GVD has to be taken into account when designing for a GVD compensation, but the losses should also be considered. To find the optimal parameters for both GVD and the losses of the system, a full set of simulation were done by varying independently all the relevant design parameters of the waveguide.

The results of simulation of the GVD and losses of the waveguide are shown in Fig. 3 when changing the thickness of the cl5 layer (Fig. 3a)), changing the thickness of cl4 layer (Fig. 3b)), and finally when changing the doping concentration of the cl4 layer (Fig. 3c)). From this plot general tendencies of the GVD of the system can be drawn. If the highly doped plasmonic-waveguide layer is getting closer to the waveguide, the dispersion of the system decreases, at the cost of increased losses (Fig. 3a)).

This phenomenon is due to the fact that the waveguide mode interaction with the plasmonic waveguide increases with decreasing distance between the two. The other important phenomenon that is observed is the fact that a thinner plasmon-waveguide layer results in a stronger coupling of the laser mode. Hence, the GVD of the system decreases in a more dramatic manner, and so the losses increase (Fig. 3b)). A similar behavior is observed when increasing the doping of the plasmon-waveguide layer (Fig. 3c)). By utilizing these three phenomena, we were able to design a top cladding arrangement that allows to have moderately small GVD while increasing the losses less than ∼50 % (below 1.1 cm⁻¹). In Fig. 3, the black dashed lines indicate the values of the losses and the GVD for the central wavenumber of 1650 cm⁻¹, the final design of the top claddings can be found in Table 1.

<table>
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<th>Parameter</th>
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<td>0.5e19</td>
<td>2.8e19</td>
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Table 1. List of parameters in the design of the waveguide. First column "Value" are the values of the optimized design, second and third columns are the minimum and maximum values for the simulations (see Fig. 3 and Fig. 4). cl4, cl5, ridge width, and ridge height correspond to the parameters explained in Fig. 2.
Fig. 3. Simulated material and waveguide GVD (left column) and losses (right column) of the QCL waveguide as a function of thickness of the layer between active region (AR) and highly doped layer (a), thickness of the highly doped layer (b), doping concentration of the highly doped layer (c).

Fig. 4 summarizes the findings by plotting the GVD and optical losses at 1650 cm$^{-1}$ as a function of the various design parameters (keeping all the other parameters fixed). In this figure, we present the evolution of the dispersion of the waveguide at 1650 cm$^{-1}$ when modifying the dimensions of the waveguide core (Fig. 4a,b), modifying the distance between highly doped
layer and active region (Fig. 4c)), modifying the thickness of the highly doped layer (Fig. 4d)), and finally modifying the doping concentration of the highly doped layer (Fig. 4e)).

![Graphs showing simulated material and waveguide GVD values for 1650 cm$^{-1}$ as a function of the waveguide facet dimensions (a and b), thickness of the layer between active region (AR) and highly doped layer (c), thickness of the highly doped layer (d), doping concentration of the highly doped layer (e).]

3. Waveguide design

The Quantum Cascade Laser (QCL) is based on a strain-balanced heterocascade active region. The gain stages consist of medium (∼ 0.5%) strained In$_{0.605}$Ga$_{0.395}$As/Al$_{0.566}$In$_{0.434}$As alloys, with a bandstructure design similar to Ref. [32]. In order to obtain a broad emission spectrum
centered at 1650 cm\(^{-1}\), the first 18 stages are designed for emission at 1620 cm\(^{-1}\) and the remaining 18 stages for emission at 1680 cm\(^{-1}\). The 4.5 mm-long lasers are epitaxial-side-up mounted, with ridge widths from 2.8 \(\mu\)m to 5.5 \(\mu\)m with uncoated facets. The emission spectrum is centered at 1650 cm\(^{-1}\), in excellent agreement with the design goal, with a maximum bandwidth of more than 80 cm\(^{-1}\) in Continuous-wave (cw) and 135 cm\(^{-1}\) in pulsed mode. CW LIV curves of a 5.5 \(\mu\)m-wide laser at various temperatures are plotted in Fig. 5. The laser exhibits a maximum output power of 520 mW at -20 °C, and 300 mW at 50 °C. The temperature coefficients of the threshold current density and slope efficiency are \(T_0 = 170\) K and \(T_1 = 265\) K, respectively. The experimental setup to characterize the comb operation is described in Fig. 6.

Fig. 5. Continuous-wave LIV curves for a 5.5 \(\mu\)m-wide laser at temperatures from -20 °C (blue) to 50 °C (red).

Fig. 6. Experimental setup of the comb measurements. The QCL output beam is collimated with off-axes parabolic mirror, attenuated with a polarizer, and sent either to thermal powermeter for optical power measurements, or to conventional FTIR for optical spectrum measurements. The QCL laser is driven with low-noise conventional current source, the RF component of the electrical signal is extracted through a bias-T and sent to a RF analyser.
The 5.5 \( \mu \)m-wide QCL is mounted on a laboratory laser housing that is capable of controlling the temperature from -30 \(^\circ\)C to more than 50 \(^\circ\)C with an integrated Peltier cooler. The driving current is sent through a bias-T, and the RF component of the current of the system is extracted from the AC port of the bias T, that signal is sent to RF analyser. The experiments were performed at temperatures from -10 \(^\circ\)C to 50 \(^\circ\)C with a step of 10 \(^\circ\)C. The optical beam is attenuated with a polarizer, and then either sent to FTIR for optical spectrum measurement, or to thermal powermeter for optical power measurements (see Fig. 6).

Frequency comb operation is observed from slightly above the threshold current up to the maximum current at all temperatures (-10 \(^0\)C to 50 \(^0\)C), with sharp peak at the intermode beat frequency of \(\sim 10.1\) GHz (corresponding to waveguide length of 4.5 mm). The linewidth of the beatnote frequency is typically less than 1 kHz, it varies with operational current from 520 Hz to 2.1 kHz, however, a significant part of the linewidth increase arises from the temperature drifts of the beat note due to the temperature controlling system (since the increased linewidths are observed at the highest operational currents where the temperature control is less stable). When operating in high phase noise regime, the linewidth of the beat note is typically more than 10 kHz. A sharp peak at the intermode beat frequency, along with the wide optical spectrum has been shown to be an indicator of a frequency comb operation for QCLs (assuming that the dispersion of the system is close to 0 fs\(^2\)/mm) [33–36]. Fig. 7 presents the comb map of this laser, showing all the measured beat note frequencies for all the temperatures. The repetition rate of the laser can be tuned by more than 100 MHz with a tuning rate of \(\sim 85\) kHz/mA.

The evolution of optical spectrum and intermode beat note RF spectrum with varying current is plotted in Fig. 8a). There are sharp dips in the spectrum due to the atmospheric absorption. The intermode beatnote signal to noise ratio is increasing with increasing driving current, since the bandwidth of locked optical modes increases (see Fig. 8b)).
Fig. 8. a): Electrically detected intermode beat note (left column) and optical spectra detected with FTIR (right column) for 5.5 μm-wide QCL for currents from 0.45 A to 1.1 A with a step of 50 mA at -10 °C. At 0.45 A the laser is operating in a single mode regime, so there is no intermode beatnote signal (top left). The intermode beat note measurement is done with span of 200 kHz, resolution bandwidth of 3 Hz, and shows a signal to noise ratio more than 60 dB. b): Signal to noise ratio (SNR) of the intermode beatnote as a function of the driving current, operating temperature is -10 °C.
A zoom on image of the optical and RF spectrum at 1 A is presented in Fig. 9, along with the atmospheric absorption spectrum taken from HITRAN database. 60 dB signal to noise ratio is observed at the sharp RF spectrum with no significant side peaks, indicating a low-noise operation of the comb [33–36].

Fig. 9. Optical spectrum detected with FTIR (top) and electrically detected intermode beat note (bottom) for a 5.5 μm-wide QCL for current of 1 A and temperature of -10 °C. The pink line at the top indicates the atmospheric absorption spectrum taken from HITRAN. The intermode beat note measurement is done with span of 200 kHz, resolution bandwidth of 3 Hz, and shows a signal to noise ratio of 60 dB.

Further analysis is performed for different ridge width devices (from 2.8 μm to 5.5 μm) with same length of 4.5 mm, all of which exhibit frequency comb operation. Fig. 10 presents the optical spectra of the various ridge width devices under the same operation voltage of ~12.5 V. The optical spectral bandwidth reduces from ~80 cm⁻¹ to ~50 cm⁻¹ with narrower ridge widths. The reduction of the optical spectral bandwidth is, to our belief, arising from the poor dispersion properties of the narrow ridge width devices (Fig. 4a)).
Fig. 10. Optical spectra detected with FTIR of 4.5 mm-long lasers with different ridge widths at the same operational voltage of 12.5 V and at -10 °C. The ridge widths vary from 2.8 µm (blue curve) to 5.5 µm (red curve).

The range of stable comb operation is also varying with ridge widths, narrower ridge width lasers exert smaller range of stable comb operation. Fig. 11 compares the stable comb operation ranges for 4.5 mm-long QCLs with different ridge widths.
Fig. 11. Frequency comb operation map of 4.5 mm-long QCL combs with different ridge widths. a): 2.8 µm, b): 3.7 µm, c): 4.6 µm, d): 5.5 µm.

Fig. 12 presents the measured optical bandwidth with output power for lasers with different ridge widths in comb operation. For the top cladding design presented in this paper, the best devices with high output power and wide spectral bandwidth are the devices with ridge widths bigger than 4.5 µm.
4. Conclusion

QCL frequency combs at ~ 6 µm have been presented. For achieving a frequency comb operation, dispersion compensation with a plasmonic-waveguide method has been used. As a result, frequency comb operation is observed slightly above the threshold current for all set temperatures.

6 µm spectral region is very crucial for various nucleic acids dynamical analysis, the development of frequency combs at this region will open novel possibilities for protein research [30, 31].

The lasers emission spectrum is centered at 1650 cm$^{-1}$, with a maximum bandwidth of 80 cm$^{-1}$. The laser exhibits a maximum output power of 520 mW at -20 °C, and 300 mW at 50 °C. The temperature coefficients of the threshold current density and slope efficiency are $T_0 = 170$ K and $T_1 = 265$ K, respectively. The noise level of the comb is tested to be low and acceptable for most of the applications such as dual-comb spectroscopy. Profound simulations have been performed of the waveguide with different top-cladding designs in terms of dispersion and losses of the waveguide. Based on these simulation results, general tendencies of the dispersion and the losses are discovered, that are useful guidelines for designing QCL combs. The presented results indicate that plasmonic-waveguide is a reliable method for achieving low dispersion in QCL waveguides while keeping losses low, and for reaching a frequency comb operation. This method can be used for potentially designing QCL combs at wavelengths down to ~ 4 µm.

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Disclosures
The authors declare no conflicts of interest.

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