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Asynchronous Upconversion Sampling of Frequency Modulated Combs

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In recent years, optical frequency combs with a frequency modulated output have attracted considerable attention. While first observed decades ago in externally modulated lasers, a whole class of semiconductor lasers has since been identified to spontaneously enter such a regime of self frequency modulation. However, the temporal properties of these sources could so far only be measured indirectly via phase-resolved frequency domain measurements. In this work, an asynchronous optical sampling technique which operates directly in time domain is demonstrated. On the basis of a mid-infrared quantum cascade laser frequency comb, both its instantaneous intensity and optical frequency are measured. The results confirm a quasi-constant intensity output alongside a close to linear frequency chirp, in accordance with phase-resolved spectral measurements. Contrary to previous works, stable phase-locking is achieved in a regime of positive intracavity dispersion resulting in an inversion of the observed frequency modulation.

1. Introduction

Optical frequency combs (OFCs)[1] are coherent sources whose spectrum consists of a set of discrete, equally spaced spectral modes. Following Fourier's theorem such spectra are produced by any periodic amplitude modulation in time domain, with the most prominent example being a regular train of ultrashort pulses. On the other extreme, also periodic frequency modulated light fields associated with a constant intensity give rise to OFCs. In both cases, it is the optical repetition period which determines the inverse optical line spacing of the spectrum.

Frequency modulated combs have been observed in a multitude of laser systems. In the early 1960's, shortly after the demonstration of the first laser,[2] signatures of frequency modulation have been reported from electro-optically modulated He–Ne lasers.[3] From the analysis of emission spectra, which revealed spectral envelopes following Bessel functions, it was concluded that the instantaneous frequency undergoes a sinusoidal modulation.[4] These early works shared the common difficulty that no means of directly measuring the temporal output were at hand due to the absence of high peak intensities in the emitted fields.

It took several decades until novel semiconductor laser sources were identified to have a similar predominantly frequency modulated output. This included quantum cascade,[5] interband cascade,[6] quantum dot,[7] and quantum dash[8] lasers, but also conventional diode lasers.[9,10] Contrary to earlier observations, these frequency modulated sources were self-starting, without the need of having an intracavity phase modulator. With the emergence of these sources also temporal characterization techniques experienced a rebirth.

Coherent beatnote interferometry (SWIFT),[11] an all-linear phase-resolved spectroscopic method, evolved to be the method of choice for characterizing frequency modulated OFCs. Originally designed for coherence measurements, it also allows to deduce temporal emission profiles. However, such time domain reconstruction relies on performing cumulative sums of spectral phase differences. As a result, SWIFT is inherently more sensitive to measurement noise than techniques capturing absolute phases. These limitations are particularly pronounced for frequency combs featuring spectral holes in the emission spectrum.[12]

Optical heterodyne techniques provide an alternative to SWIFT. On the one hand, dual-comb detection schemes[13] allow direct access to absolute spectral phases and amplitudes but require a reference frequency comb of known complex spectrum. On the other hand, in stepped heterodyne techniques,[14] a single mode laser of arbitrary spectral phase is sufficient for the measurement of spectral amplitudes and phase-differences. However, in this case the single mode laser must be widely tunable covering the full spectrum of the frequency comb.

Apart from aforementioned methods which rely on frequency domain measurements, optical sampling techniques permit to measure light fields directly in time domain. This approach has proven particularly successful in the terahertz frequency range,[15] but can also be applied to shorter wavelength ranges

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provided that the sampling pulses are considerably shorter than the time scales of the field to be measured.

In this article, we use such an optical sampling technique to measure both the instantaneous intensity and frequency of a frequency modulated OFC. Near-infrared ultrashort pulses are employed to asynchronously sample the intensity of a mid-infrared quantum cascade laser (QCL) frequency comb.[5,16] With the help of a tunable optical bandpass filter, we additionally acquire a spectrogram of one repetition period of the comb. The obtained results are compared with phase-resolved spectral measurements.

2. Experimental Section

Sum-frequency generation (SFG) provides the means to cross-correlate light fields. In particular, it can serve as an optical sampling mechanism when used in combination with ultrashort laser pulses, which act as rapid gates in temporal domain. Here, the situation where a femtosecond mode-locked laser (MLL) was used to coherently sample the output of a QCL frequency comb using SFG was considered.

In such a process, the generated sum-frequency signal was proportional in intensity to the intensity product of the two incoming light fields I_{QCL} and I_{MLL}. Given the ultrashort pulse length of the MLL as compared to the QCL repetition period ($\tau_{\mathrm{MLL}} \ll T_{\mathrm{QCL}}$), the intensity of the MLL could be approximated as a periodic train of δ-functions.[15] Taking into account the frequency comb nature of the QCL, it was found for the SFG intensity:

$$\begin{aligned} I_{\mathrm{SFG}}(t) &\propto I_{\mathrm{QCL}}(t) \times I_{\mathrm{MLL}}(t) \\ &\propto \left| \sum_n A_n e^{2\pi i (f_{\mathrm{ceo}}^{\mathrm{QCL}} + n f_{\mathrm{rep}}^{\mathrm{QCL}}) t} \right|^2 \times \sum_{m=-\infty}^{\infty} \delta\left(t - \frac{m}{f_{\mathrm{rep}}^{\mathrm{MLL}}} \right) \end{aligned} \tag{1}$$

where A_n are the QCL's complex modal amplitudes, while $f_{\mathrm{rep}}^{\mathrm{QCL}}$ and $f_{\mathrm{rep}}^{\mathrm{MLL}}$ represent the repetition frequencies of the QCL and MLL, respectively. The QCL carrier offset frequency was denoted by $f_{\mathrm{ceo}}^{\mathrm{QCL}}$.

By expanding I_{MLL} into its Fourier components, Equation (1) could be brought to the following form

$$\begin{aligned} I_{\mathrm{SFG}}(t) &\propto \sum_{n,n'} A_n A_{n'}^* e^{2\pi i (n-n') f_{\mathrm{rep}}^{\mathrm{QCL}} t} \times \sum_{m=-\infty}^{\infty} e^{2\pi i m f_{\mathrm{rep}}^{\mathrm{MLL}} t} \\ &\propto \sum_{n,n'} A_n A_{n'}^* e^{2\pi i (n-n') \Delta f t} \times \sum_{m'=-\infty}^{\infty} e^{2\pi i m' f_{\mathrm{rep}}^{\mathrm{MLL}} t} \\ &\propto \left| \sum_n A_n e^{2\pi i n \Delta f t} \right|^2 \times \sum_{m'=-\infty}^{\infty} \delta\left(t - \frac{m'}{f_{\mathrm{rep}}^{\mathrm{MLL}}} \right) \\ &\propto I_{\mathrm{QCL}}^{\Delta f}(t) \times I_{\mathrm{MLL}}(t) \end{aligned} \tag{2}$$

with $\Delta f \equiv f_{\mathrm{rep}}^{\mathrm{QCL}} - k \times f_{\mathrm{rep}}^{\mathrm{MLL}}$ and $k \equiv \mathrm{int}(f_{\mathrm{rep}}^{\mathrm{QCL}} / f_{\mathrm{rep}}^{\mathrm{MLL}})$. In this equation, $I_{\mathrm{QCL}}^{\Delta f}$ is a slowly-varying replica of the original QCL waveform I_{QCL} where the repetition rate $f_{\mathrm{rep}}^{\mathrm{QCL}}$ has been replaced by Δf.

In an experiment, $I_{\mathrm{QCL}}^{\Delta f}(t)$ could be directly obtained by acquiring $I_{\mathrm{SFG}}(t)$ at a sampling rate of $f_{\mathrm{rep}}^{\mathrm{MLL}}$. If additionally the reference signal $I_{\Delta f}(t) \propto \cos(2\pi \Delta f t + \phi_0)$ was acquired, each sampling point t could be mapped to its respective optical waveform position $t' = \xi(t)$

$$\begin{aligned} \xi : \quad & (-\infty, +\infty) \to [0, T_{\mathrm{rep}}^{\mathrm{QCL}}) \\ & t \mapsto \frac{\arg\left(I_{\Delta f}(t) \right)}{2\pi f_{\mathrm{rep}}^{\mathrm{QCL}}} \end{aligned} \tag{3}$$

The phase offset ϕ_0 was of subordinate physical relevance, as it only introduced a constant time delay in the reconstructed intensity waveform. It was to be noted that Δf should not be a rational fraction of $f_{\mathrm{rep}}^{\mathrm{QCL}}$, as otherwise, depending on the bandwidth of $I_{\mathrm{QCL}}^{\Delta f}$, aliasing effects might occur (see Section S5, Supporting Information, for details).

The general sampling procedure is summarized in **Figure 1**a. Ultrashort laser pulses from the MLL asynchronously sweep the repetition period of the QCL comb, with subsequent pulses separated by $\tau = \Delta f / (f_{\mathrm{rep}}^{\mathrm{QCL}} f_{\mathrm{rep}}^{\mathrm{MLL}})$. Corresponding intensity samples, in the figure denoted with circles, then allowed for a full intensity reconstruction using Equation (3). This technique was called asynchronous upconversion sampling (ASUPS).[12]

The experimental configuration is shown in Figure 1b. For feedback suppression, an optical isolator was placed at the output of the QCL. The beam was then guided through a tunable optical bandpass filter. It was arranged in a retro-reflective geometry, incorporating a blazed grating, a mirror and a lens. The lens was separated from the grating and mirror by its focal length to prevent introducing residual group delay dispersion (GDD) to the light field. A movable slit close to the retro-reflector enabled spectral bandpass filtering.

The QCL with a center wavelength around 8 µm was then spatially overlapped with a 1.55 µm MLL emitting ultrashort pulses with a pulse duration of $\tau_{\mathrm{MLL}} \approx 100$ fs. The two light fields nonlinearly interacted inside an $AgGaS_2$ crystal to generate a SFG signal around 1.3 µm, as shown in Figure 1c. The chosen crystal length of 0.4 mm is a trade-off between efficient frequency conversion over the full bandwidth of the involved laser fields while maximizing the overall sum-frequency signal (details in Sections S1 and S2, Supporting Information). As collinear type-I phase matching was employed, a polarizing beam splitter and bandpass filters were used to separate the SFG signal from the two pump beams at 8 and 1.55 µm. The remaining signal was subsequently measured on an avalanche photodiode (APD) and acquired at the sampling rate $f_{\mathrm{rep}}^{\mathrm{MLL}} \approx 90$ MHz, as obtained from an internal photodiode of the MLL. A phase shifter was used to sample on the peak of each pulse arriving on the APD.

The reference signal $I_{\Delta f}(t)$ was simultaneously acquired on a separate channel. In the experiment, $f_{\mathrm{rep}}^{\mathrm{QCL}} \approx 12$ GHz was injection locked to an external stabilized microwave source. By electrically mixing $f_{\mathrm{rep}}^{\mathrm{QCL}}$ with the high beat tone $k \times f_{\mathrm{rep}}^{\mathrm{MLL}}$ (measured on a fast photodiode) and filtering the product with a low-pass filter of bandwidth $f_{\mathrm{rep}}^{\mathrm{MLL}}/2$, $I_{\Delta f}(t)$ could be directly accessed as shown

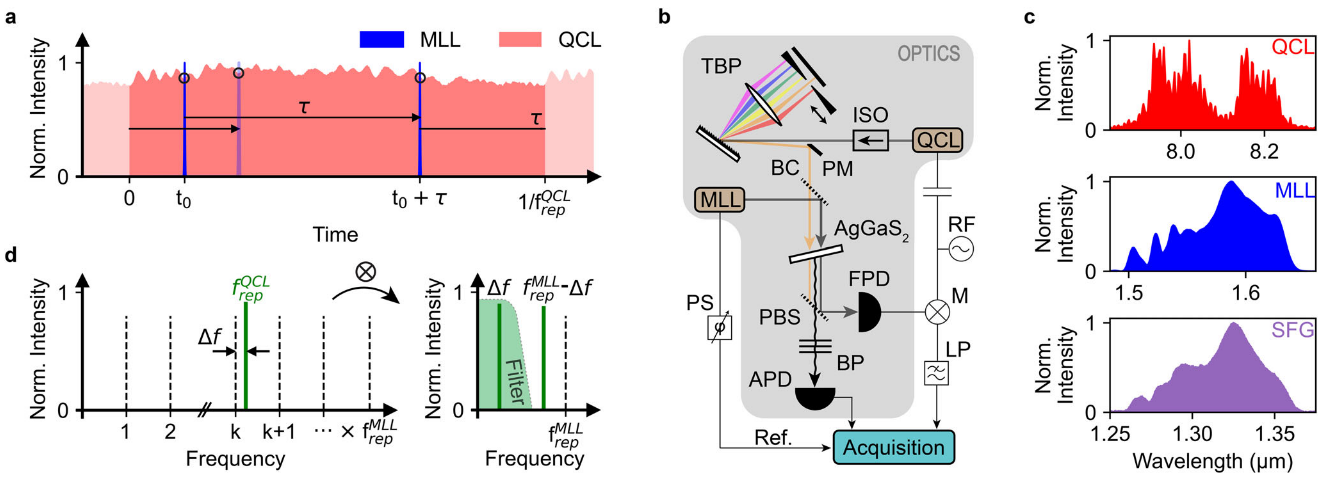

Figure 1. Asynchronous upconversion sampling. a) An MLL is used to asynchronously sample the output of a QCL frequency comb. When projected onto a single QCL period, individual waveform samples are separated by τ. b) Optical sampling is accomplished using SFG in $AgGaS_2$. TBP: tunable optical bandpass filter, ISO: optical isolator, PM: pick-off mirror, BC: beam combiner, RF: radio frequency synthesizer, FPD: fast (≈ 15 GHz) photodiode, M: frequency mixer, PS: phase shifter, PBS: polarizing beam splitter, LP: low pass filter, BP: optical bandpass filters, APD: avalanche photodiode. c) Spectra of the three waves involved in the SFG process. d) In order to assign each intensity sample to its respective waveform position, the frequency difference Δf is additionally acquired in the experiment. It is obtained by electrically mixing $f_{\mathrm{rep}}^{\mathrm{QCL}}$ with $k \times f_{\mathrm{rep}}^{\mathrm{MLL}}$ and applying a lowpass filter.

schematically in Figure 1d. With this signal and after coherent averaging (see Section S3, Supporting Information, for details), $I_{\mathrm{QCL}}^{\Delta f}(t')$ could then be reconstructed.

3. Results

In this work, we employ a 3.75 mm long QCL comb with a plasmon-enhanced waveguide for dispersion compensation. The device is high-reflection coated at the back facet and cooled to −10 °C using a Peltier element. At an operation current of 1 A, we measure an average output power of 760 mW. The emission spectrum is centered around 1285 cm^{-1} with a spectral bandwidth of ≈ 40 cm^{-1}.

Figure 2a shows the measured instantaneous intensity as obtained by ASUPS for the full QCL spectrum. We observe a quasi-constant intensity, which, considering above mentioned spectral bandwidth, indicates a strong frequency modulation of the emitted field. The waveform is recorded with a temporal resolution of ≈ 200 fs, limited by the finite MLL pulse length and timing jitter of the sampling pulses (see Section S4, Supporting Information, for details).

The corresponding instantaneous frequency is measured using the tunable optical bandpass filter shown in Figure 1b. The slit width is set such that a spectral fraction of ≈ 5 cm^{-1} is transmitted through the filter. Then, the slit is step-wise moved across the full spectrum of the QCL giving rise to pulses of varying group delay. Figure 2a exemplarily shows such a pulse as measured by ASUPS. For each pulse, its center wavenumber is measured using a Fourier transform spectrometer. Figure 2c summarizes the measured center wavenumber as a function of the slit position.

A spectrogram, indicative for the instantaneous frequency of the light field, can then be generated as shown in Figure 2b. We observe a monotonic frequency modulation, where the frequency increases close to linearly with time. Within one repetition period, the laser sweeps exactly its full spectral bandwidth.

When performing the measurement, particular care was taken not to introduce residual GDD at the spectral filter. Figure 2d shows the temporal waveform as recorded using ASUPS with and without the optical filter inserted into the beam path. With the filter inserted and the slit fully open, no considerable change in temporal intensity can be observed, from which we conclude the filter GDD to be considerably smaller than the GDD inherent to the light field.

For reference, we performed phase-resolved spectral measurements[11] on the same device. Figure 2e,f displays the obtained instantaneous intensity and frequency, respectively. In close agreement with ASUPS, we observe a quasi constant intensity associated with a positive frequency chirp. The evaluated spectral coherence is shown in Figure 2g confirming a close to unity degree of mutual coherence among comb lines over the full spectral range. For further experimental details please refer to our previous publication.[12]

4. Discussion

The results shown in Figure 2 unequivocally demonstrate that QCL combs emit a close to maximally frequency modulated field. The two fundamentally different measurements performed in this work, relying on temporal and spectral averaging respectively, are in excellent agreement. They reveal a linear frequency chirp which exactly sweeps the laser spectrum within one repetition period.

The full coherence of the emitted waveform is confirmed by the vanishing intensity background in the ASUPS measurements reported in Figure 2b. This observation is in excellent agreement with the spectrum product and SWIFT spectrum in Figure 2g which are commensurate over the full spectral range. It is to be mentioned that the latter is no formal proof of comb

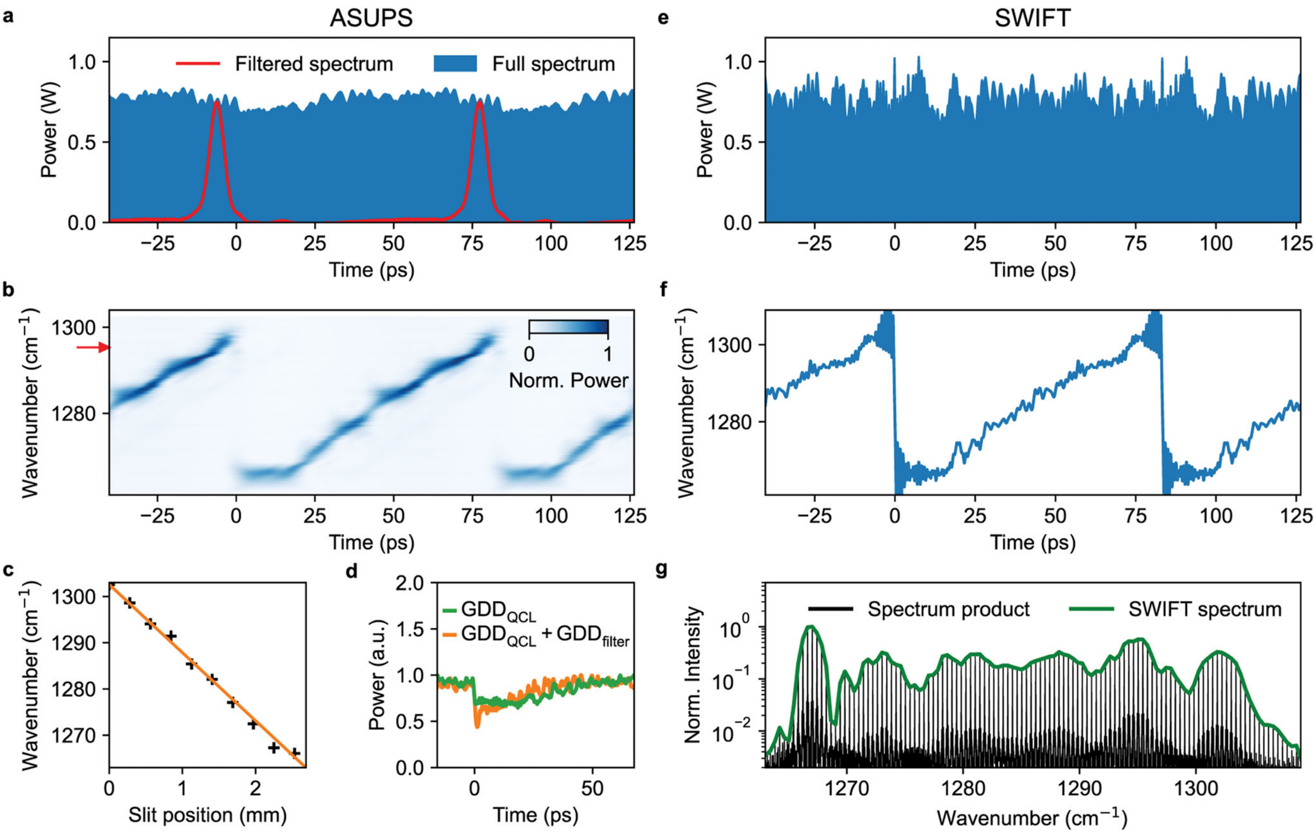

Figure 2. ASUPS and SWIFT measurements of a mid-infrared QCL frequency comb. a) Instantaneous intensity as measured by ASUPS for the full and spectrally bandpass filtered spectrum. b) Instantaneous frequency as measured by ASUPS using a tunable optical bandpass filter. The red arrow indicates the waveform plotted in red in the previous subfigure. c) Center wavenumber as transmitted through the optical bandpass filter for different slit positions. d) Instantaneous intensity of the QCL as measured without (GDD_{QCL}) and with ($GDD_{QCL} + GDD_{filter}$) the grating filter. e,f) Instantaneous intensity and frequency as measured by SWIFT. g) Comb coherence as measured by SWIFT.

coherence, as the proportionality constant relating the two quantities is not readily accessible by experiment and would require a sophisticated calibration.

The physics behind frequency modulated OFCs can be understood in the framework of the generalized nonlinear Schrödinger equation (GNLSE).[17] In this picture, the frequency modulated nature arises from a predominantly phase-dependent potential term, physically originating from cross-steepening nonlinearities. The GNLSE can be solved analytically, resulting in a linearly frequency chirped solution with the instantaneous frequency proportional to the inverse group velocity dispersion β^{-1}. These non-dispersive solutions must span a full cavity round trip in order to form a stable solution.

From this model several implications can be deduced. First, if the frequency modulated state extends over a bandwidth larger than supported by the gain medium, the frequency comb destabilizes. In particular, for $\beta = 0$ phase-locking is considered impossible, as it would require an infinite gain bandwidth.

Second, away from this region of instability around $\beta = 0$ there are two regions where stable phase-locking is expected. These regions are found in the positive ($\beta > 0$) and negative ($\beta < 0$) dispersion ranges and manifests themselves in maximally chirped solutions with a positive and negative frequency slope, respectively.

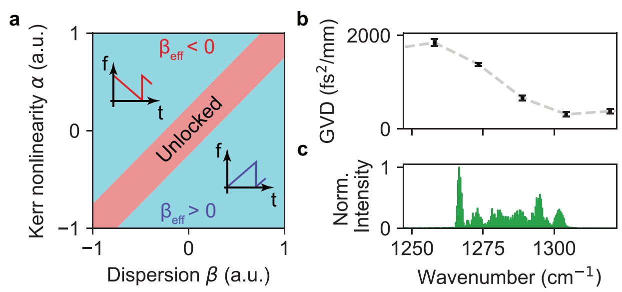

Figure 3. QCL comb formation and dispersion. a) Stable phase-locking is only possible in a regime of strictly positive or negative effective dispersion β_{eff}, while in the region around $\beta_{eff} = 0$ the laser remains unlocked. b) GVD as measured using the Hakki–Paoli technique below lasing threshold. c) Spectrum of the QCL used in this work.

Third, Kerr nonlinearities α are not required for comb formation but have the effect of shifting the effective dispersion β_{eff} and therefore the range over which frequency combs can form. **Figure 3**a schematically summarizes above mentioned points.

In this context, we can interpret the results shown in Figure 2. We observe a field which is chirped close to linearly from the red to the blue part of the spectrum. This can be readily understood by means of Figure 3a, where a positive frequency chirp is expected in the stability region with $\beta_{eff} > 0$.

While β_{eff} is not easily accessible by experiment, we have measured β employing the Hakki-Paoli method.[18] The results are shown in Figure 3b. Over the full laser spectrum (Figure 3c) we observe a positive group velocity dispersion (GVD). Taking into account reasonable values for the Kerr nonlinearity,[19] we conclude that the laser operation point indeed lies in a region of parameter space where a positive frequency chirp is expected. Similarly, previous measurements on QCL combs, where the frequency modulation was inverted, can be ascribed to a region of negative GVD.[20,21]

5. Conclusion

In conclusion, we have applied ASUPS, an asynchronous optical sampling technique, for the temporal characterization of QCL combs. Our results show that QCL combs are strongly frequency modulated with the field exactly sweeping the laser bandwidth within one repetition period. The observed frequency chirp is close to linear, in accordance with phase-resolved spectral measurements. We attribute the sign of the observed frequency chirp to the measured positive laser dispersion.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

P.T. conducted the experiments, performed the data analysis, and wrote the manuscript. A.F. contributed to the SWIFT data analysis, and M.B. performed the initial characterization (light–current–voltage characteristic, spectrum) of the device used in this work. F.K. processed the QCL with the semiconductor heterostructure grown by M.B., and J.F. supervised this work.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

frequency combs, frequency modulation, mid-infrared lasers, optical sampling, quantum cascade lasers

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