

Tuning properties of mid-infrared Fabry-Pérot quantum cascade lasers for multiheterodyne spectroscopy

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Abstract—Injection current tuning properties of an 8.5 μm Fabry-Pérot mid-infrared quantum cascade laser are evaluated by analyzing the mode-by-mode frequency tuning behavior with an identification of high-noise regimes in a delayed self-heterodyne experiment. We find that modes on the edges of the spectral envelope exhibit anomalous tuning coefficients compared to those in the center. Furthermore, the frequencies of individual modes are susceptible to parasitic etalons, likely causing laser frequency pulling. Despite the complicated tuning behavior, low phase-noise operating regimes exist, and are compatible with high resolution multiheterodyne spectroscopy of gases.

Since the first demonstration in 1994 [1], the quantum cascade laser (QCL) has become an increasingly important coherent semiconductor light source for optical gas spectroscopy in the mid-infrared spectral region. This region is of particular interest since many environmentally important molecules exhibit their largest absorption cross sections in this wavelength range. Although the QCLs were initially confined to cryogenic operation, which hindered in-field applications, the rapid developments quickly rendered stable room-temperature devices, subsequently making them well-suited for various practical sensing applications. Early research efforts were primarily focused on the development of spectrally pure, single-mode, distributed feedback (DFB) QCLs compatible with tunable diode laser absorption spectroscopy (TDLAS) applications. However, in recent years an increasing interest in broadband Fabry-Pérot (FP) devices operating in frequency comb, and quasi-frequency-comb regimes has emerged. In 2012 Hugi *et al.* demonstrated that in selected current regimes the FP-QCLs possess a sufficiently low intracavity dispersion, which enables the formation of stable frequency combs due to four wave mixing (FWM) [2]. As a result, broadband dual comb spectroscopic techniques that utilize beating between two optical frequency combs with different repetition rates can be realized with compact FP-QCL devices [3]. To ensure QCL comb operation over a broad frequency range, dispersion engineering of the active region or external compensation by Gires-Tournois

interferometer (GTI) mirrors have been successfully employed [4]. While, such techniques reduce the intermode beatnote linewidth down to sub-kilohertz levels, they require additional design steps and custom fabrication of the structures.

Undoubtedly, stable operation of the lasers is desired, however, for applications that do not require sub-kilohertz linewidths free-running, non-comb optimized QCLs are a viable light source for high resolution multiheterodyne spectroscopy (MHS) [5]. One such application is gas spectroscopy at atmospheric pressures, where absorption linewidths are in the gigahertz range and high-resolution spectra can be reliably measured via modulation of the laser injection current or laser temperature [3]. In such spectrometers, accurate knowledge of the mode tuning characteristics is critical for proper system operation, thereby making careful tuning characterization of sources of large importance.

In this work, a FP-QCL emitting coherent mid-infrared radiation centered at 1180 cm^{-1} with a mode spacing of approximately 40.8GHz (1.36 cm^{-1}) was characterized. The laser was placed in a low pressure TEC-cooled mount and collimated with a f/0.6 germanium lens, as described in our previous works [5-6]. The QCL tuning behavior as a function of the injection current at a constant temperature of 274K was characterized by recording the optical (FTIR) and self-heterodyne (SH) spectra simultaneously, as shown in Figs. 1a÷b. In the optical spectrum measurement, a commercially available Fourier transform spectrometer (Nicolet 8700) was used, whereas the self-heterodyne experiment involved the use of an AR-coated acousto-optic modulator (Intraaction AGM-406B) driven by a high power RF amplifier (Intraaction GE-4020) at 40 MHz, two beam splitters in a ~40 cm delay branch, and a high speed mid-IR photodetector (VIGO PV-4TE-10.5) measuring the self-heterodyne beat note using a RF spectrum analyzer. As shown in Fig. 1a, one can observe a broadening of the SH beat note noise pedestal up to an injection current of approximately 180mA.

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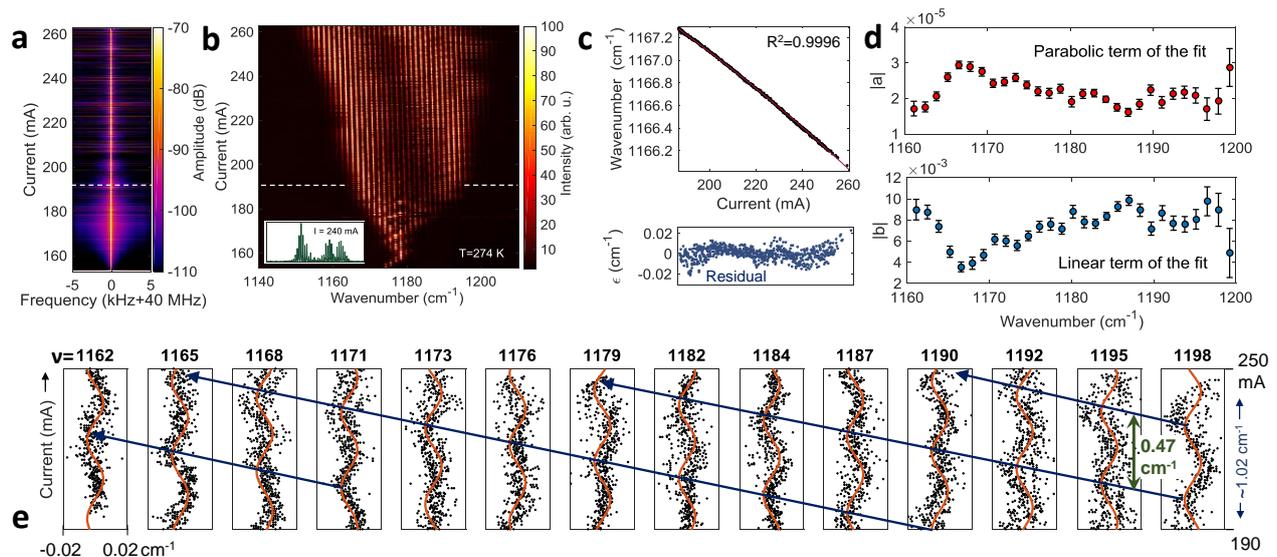


Fig. 1. Self-heterodyne (a) and FTIR measurements (b) as a function of the injection current. After fitting a second order polynomial to the retrieved position of the modes (c), one can observe a stronger nonlinearity of the modes in the dominant (left) branch of the spectrum (d). The residuals of the fit (e) (every other mode is shown) possess a periodic pattern with a FSR of approximately 0.47 cm^{-1} , indicating a slight feedback effect on the lasing frequency caused by an etalon in the system. The globally fitted fringe is plotted with solid lines together with diagonal equiphase arrows.

In this regime (marked with dashed line in Figs. 1a=b) only few modes are lasing and the broadband mode structure is slowly developing while periodically switching between single and multimode operation, as visible in Fig. 1b. When a broader mode structure is present i.e. at higher currents, the frequency noise of the SH beat note is clearly reduced (narrower beat note) with sporadic high noise regimes for certain current settings. Note that to broaden the SH beat note of a multimode laser, it is sufficient to have only one or a few modes with a higher frequency noise. There are several interferometric techniques developed to evaluate which parts of the spectrum contribute to stable coherent laser operation, such as SWIFT (shifted wave interference Fourier transform spectroscopy) [7] and intermode beat spectroscopy [2]. However, these methods require ultrafast photodetectors, such as quantum well infrared photodetectors (QWIPs). Thus the SH technique, which can only extract partial information about the source coherence but significantly simpler to implement and does not require ultrafast electronics, can be used as a rough guide for finding suitable operating regimes when the modes are spaced by tens of GHz.

Figure 1b shows the behavior of the emitted optical spectrum, where two (or more) groups of modes are formed and separated from each other as the injection current is increased. A detailed theoretical explanation of this phenomenon is given by A. Gordon *et al.* [8], where the spectral splitting is related *inter alia* to the ultrafast dynamics of the QCL gain. From a spectroscopic viewpoint, the suppressed intensity in the central part of the spectrum limits the useful spectral coverage of the

sources. While it is possible to utilize the weaker modes for MHS, they require a spectrally-matched strong local oscillator for optical heterodyning, and will inevitably suffer from a degraded signal-to-noise ratio.

To characterize the mode-by-mode tuning behavior in detail, the FTIR spectral data was resampled, followed by an estimation of the position of the peaks, which were fit to a second order polynomial, as shown in Figure 1c. The nonlinear tuning characteristics stems from a proportionality of the optical frequency of the modes to the change of the active region temperature T , which in turn is a function of thermal resistance and electric power delivered [9], all exhibiting nonlinear behavior. Despite the model simplicity, the fit remains within excellent agreement of the experimental data ($R^2 > 0.994$ for all investigated modes), albeit with a small sine-wave-like fit residual that occurs for all the modes with a noticeable phase shift. This phase shift is highlighted by the diagonal arrows in Fig. 1e. After rescaling from current to optical frequency, and fitting a squared sine to the data, the period of the pattern is estimated to be approx. 0.47 cm^{-1} .

With the assumption of uniform thermal tuning, it is expected that the linear term of frequency tuning will show a gradual increase with an increasing wavenumber while the parabolicity of the tuning curve will be preserved. However, based on the performed measurements, the tuning of the non-comb-optimized QCL used in this study, shows considerable deviations from this gradual increase in tuning rate (Fig 1d). In particular, modes in the $1160\text{--}1168 \text{ cm}^{-1}$ seem to possess anomalously high linear terms accompanied by lower parabolicity of the tuning curve as compared to the neighboring modes.

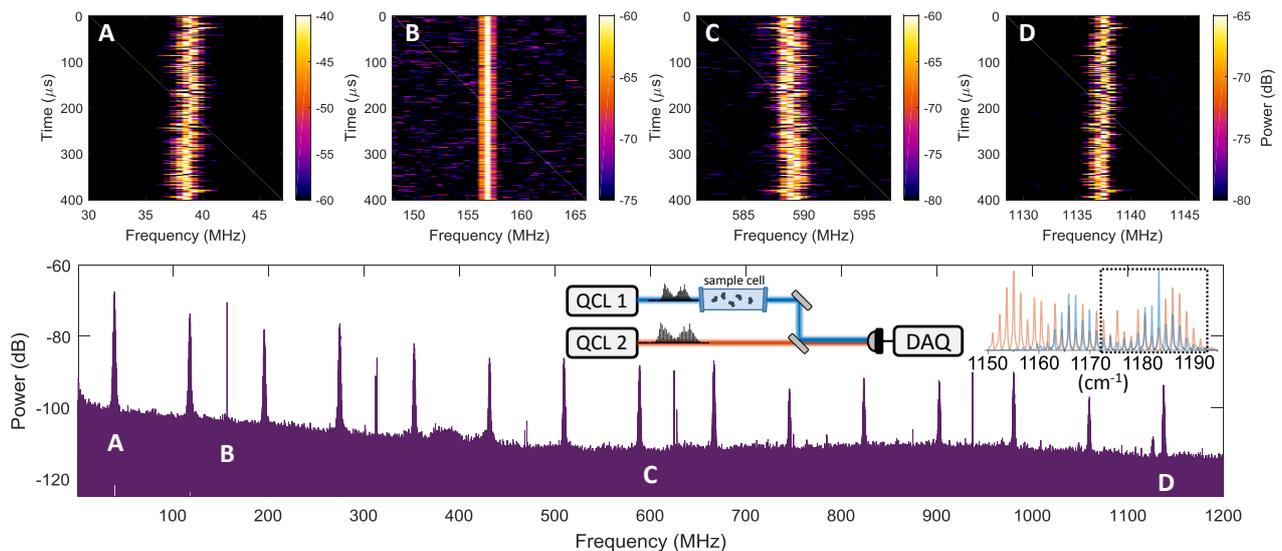


Fig. 2. Multiheterodyne spectrum acquired with a fast oscilloscope in 400 μs plotted together with periodograms of the RF beatnotes (A, B, C, D). Inset shows a simplified QCL-based multi-heterodyne spectrometer together with the spectral overlap region marked with a dashed border rectangle. Note the anti-correlation of beat note C with respect to A and D due to spectral folding. The technique allows to double the number of RF beat notes within the detector bandwidth, yielding a DC-centered spectrum with folded (aliased) and unfolded components moving in opposite directions [10].

This has been attributed to the fact that the dominant left lobe of the emission spectrum shifts its position towards lower optical frequencies. In DFB QCLs, tuning is usually strongly affected by the position of the mode with respect to the gain peak, which can be tuned thermally. Similar thermal dependence of the gain profile characteristics is expected to appear in the FP-QCLs. Additionally the periodic fit residuals have been attributed to a parasitic etalon in the system that introduces optical feedback, which causes a slight change ($\sim 0.015 \text{ cm}^{-1}$) in the lasing frequency via frequency pulling [11]. While the optical frequencies of individual modes are repeatable between subsequent power cycles, their intensities are only approximately the same. The fluctuations of the mode envelope are observed during power cycling, which is related to the complex laser dynamics of a multimode laser, but once the laser is turned on it settles into a stable mode structure that can be used for spectroscopic measurements.

Despite the complicated tuning behavior, nonlinearities, and deviation from an equidistant mode structure, the laser is perfectly suitable for MHS. To demonstrate this capability two such FP devices have been heterodyned and biased for operation in the low phase noise regime (Fig. 2 inset). Figure 2 shows a comb of RF frequencies obtained from the multiheterodyne process. Since this is an aliased multiheterodyne spectrum, same-sign beat notes will exhibit a highly correlated phase noise (A and D), whereas folded and non-folded beat notes will be anti-correlated (A or D vs C). This is also manifested by a narrow self-mixing component (B), being the result of beating between RF beat notes due to residual nonlinearities in the signal path. The tens of kHz of self-mixing linewidth indicate the ultimate spectral resolution achievable with this kind of laser source.

In conclusion, the tuning characteristics of a Fabry-Pérot QCL have been investigated. Although the optical spectrum shows significant complexity over the full bias range, it is possible to identify low phase noise (quasi-frequency-comb) regimes, where stable and equidistant multiheterodyne beat notes can be observed. These regions are found to be suitable for high resolution optical spectroscopy of gases [6].

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