Noise properties in multi-heterodyne spectrometers based on quantum- and interband-cascade lasers

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Abstract: The noise properties of two multi-heterodyne spectrometers based on multi-mode semiconductor lasers (QCLs and ICLs) are investigated to determine their detection performance limitations. Both laser technologies provide well-defined multi-heterodyne beat-note structures suitable for spectroscopic measurements.

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Spectroscopic chemical sensing based on semiconductor lasers is already an established method for highly sensitive and selective detection. Compared to other spectroscopic techniques (FTIR, UV-VIS or IR spectrometry) it provides fast measurement rates (kHz-MHz), high spectral resolution (<100 MHz) and excellent signal to noise ratio (>100 dBc/Hz). However, most semiconductor lasers have a rather narrow tuning range (< 1 %) and, hence, only molecular species with narrow transitions can be targeted. A common solution to this problem is to use external cavity lasers, which provide a much larger tuning range (> 10 %) and allow for multiple species detection at the cost of reduced tuning speed, decrease in robustness (due to mechanical moving parts), and generally higher cost.

A particularly elegant way to approach the problem of performing broadband and high resolution spectroscopic detection using lasers is the dual-comb spectroscopy technique [1]. This methodology was originally developed in the context of mode-locked frequency comb sources [2]. This field has recently received considerable attention due to possibility of replacing the frequency combs with semiconductor lasers to enable robust and cost efficient design. Since semiconductor Fabry-Perot (FP) lasers emit a comb-like frequency spectrum they are attractive alternative sources for this task. However, standard FP lasers are typically not mode-locked because of the missing intrinsic locking mechanism and non-zero group velocity dispersion inside the cavity. Nevertheless, quantum cascade lasers (QCLs) can be engineered to exhibit a behavior similar to FM locking at certain operation conditions, which can then be utilized for high-performance dual comb sensing [4]. Since design of an intrinsically mode locked laser is a specialized task and not available for all semiconductor lasers types (only QCLs provide this feature so far), the question is what are the performance limits with non-locked semiconductor FP lasers. We have already demonstrated a spectroscopic application of non-locked FP-QCLs to multi-heterodyne spectroscopy [3]. In this work we further study the performance of standard FP-QCLs and interband cascade lasers (ICLs) and their suitability to perform multi-heterodyne spectroscopic detection.

In the setup shown in Fig. 1 we have studied the noise limits that determine the precision of the spectral measurement. In transmission measurements the intrinsic fluctuation of the instantaneous beat note power (i.e., its power spectral density) has been assessed to determine amplitude noise. For stability and linewidth quantification we have measured the power spectral density of the instantaneous beat note frequency fluctuations. Furthermore, the spectrum of the squared signal in the time domain (i.e., the RF-self-mixing spectrum) is recorded to uncover correlations in the frequency noise between the beat notes.

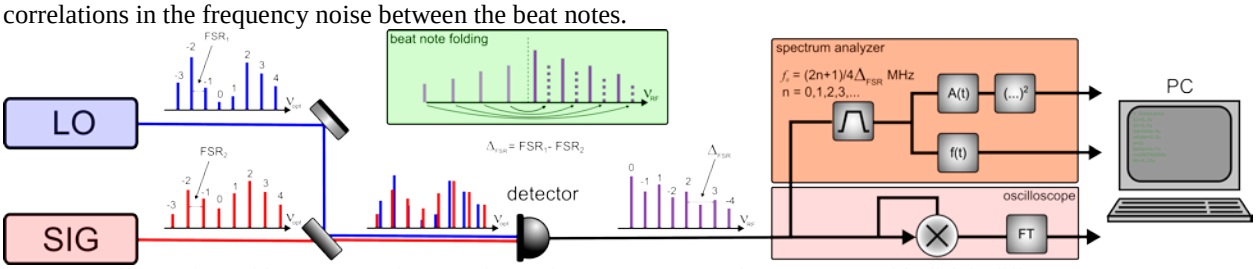

Fig. 1: The experimental layout. Two Fabry Perot laser pairs (8.5 µm QCLs and 3.5 µm ICLs) with slightly different FSR (Δ_{FSR}) are used. The optical longitudinal mode spectra are aligned at the center mode (labeled "0") with distance $\Delta_{FSR}/4$. This folds the beat notes from the negatively numbered longitudinal mode pairs interleaved with the positive ones resulting in equidistantly spaced (with $\Delta_{FSR}/2$) RF beat notes. The signals of one beat note and the full spectrum are recorded for analysis. FT: Fourier transform.

The frequency noise measured for QCLs is significantly lower than noise observed for ICLs. This is due to the low spontaneous emission and the near zero linewidth enhancement factor, which makes QCLs distinct from any other laser technology. ICLs based on interband transition exhibit frequency noise that is similar to diode lasers.

The RF self-mixing spectrum reveals hidden correlations between the beat notes (see bottom panels of Figs 2 and 3). The self-mixing spectrum contains the sum and difference frequencies of all beat note pairs. Since the distance between all beat notes with the same sign (e.g. between (0,1), (1,2), (-1,-2)) is a multiple of Δ_{FSR}, distinct frequency components are observed there. These self-mixing beat notes observed for QCLs (Fig. 2) are clearly much narrower (~15 kHz) than the beat notes in the original spectrum (1.9 MHz). This indicates that the frequency noise on the beat notes is highly correlated with an uncorrelated part of only 15 kHz. For ICLs optical heterodyne beat note width of 11 MHz was observed and the self-mixing spectrum indicates the peak width of 300 kHz which is also very promising for high resolution spectroscopy.

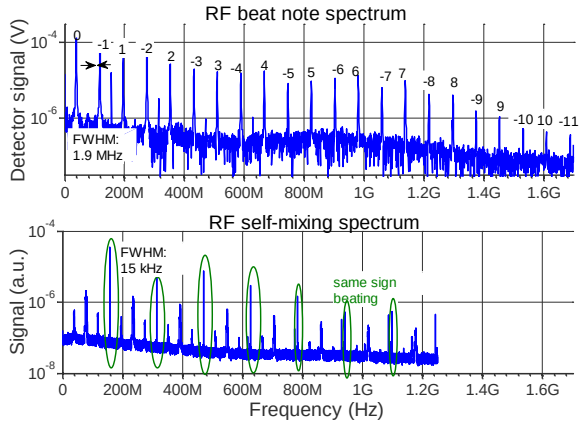

Fig. 2: Multi-heterodyne beat note spectrum for the QCLs emitting at 8.5 µm. A 21×1.4 cm^{-1} = 29.4 cm^{-1} spectral coverage with $\Delta_{FSR} \approx$ 160 MHz is obtained. Although the beat notes are ~1.9 MHz wide (top), the frequency noise is highly correlated among them, with only a 15 kHz uncorrelated contribution (bottom).

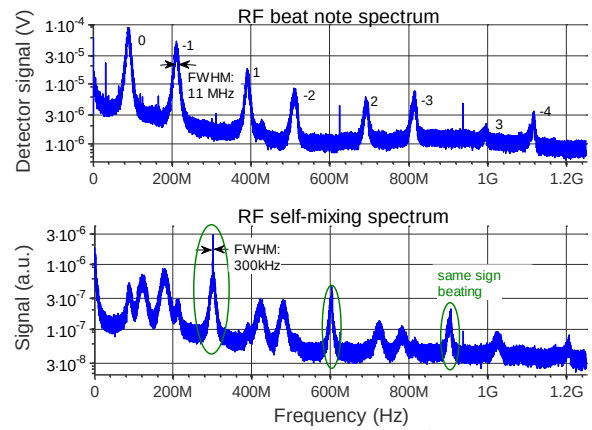

Fig. 3: Multi-heterodyne beat note spectrum for the ICLs emitting at 3.5 µm. A 8×0.95 cm^{-1} = 7.6 cm^{-1} spectral coverage with $\Delta_{FSR} \approx$ 300 MHz is obtained. The beat notes are ~11 MHz wide (top). The frequency noise seems largely uncorrelated (due to the broad peak at 300 MHz in the RF self-mixing spectrum), but with a very distinct correlated noise contribution of 300 kHz width.

The optical heterodyne beat note width is representative for a long term linewidth (due to technical noise [5]) and self-mixing beat notes indicate short term (i.e. intrinsic) linewidth due to spontaneous emission which is uncorrelated between the longitudinal modes in the laser.

Fig. 4 shows the power spectral densities of a selected beat note. QCLs show relative intensity noise floor of ~$5.7 \cdot 10^{-5}$, while ICLs approach $3.5 \cdot 10^{-4}$.

Although the intrinsic linewidth observed here for FP-QCLs is not as narrow (500 Hz) as in ref [4], an upper bound on the intrinsic linewidth of 15 kHz obtained here would be sufficient for ultra-high resolution spectroscopic applications (e.g. Lamb dip spectroscopy). Free running FP-QCLs with 1.9 MHz linewidth show an excellent potential for high resolution multi-heterodyne spectroscopy of gases.

As expected, we obtain wider beat notes for ICLs, but even with 11 MHz (~0.0004 cm^{-1}) of free-running linewidth this laser technology shows excellent potential for spectroscopy in the 3 µm region. Furthermore, the peak width of 300 kHz in the self-mixing spectrum indicates room for further improvements.

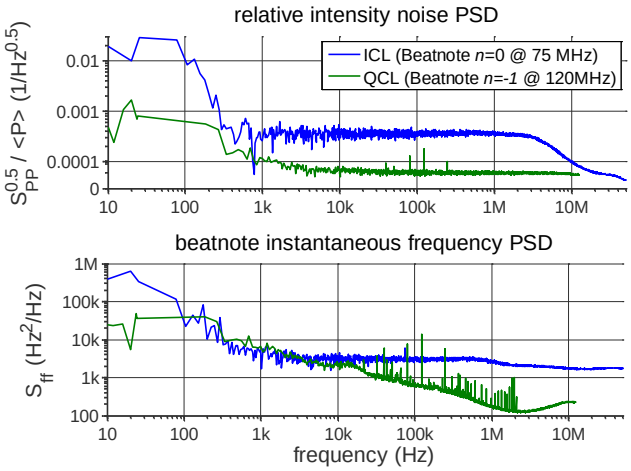

Fig. 4: The power spectral density of the instantaneous beat note power (top), which is proportional to the optical power used for measuring the transmission of a sample, and the beat note frequency noise (bottom).

In conclusion, multi-heterodyne spectroscopy with multi-mode semiconductor lasers enables a multitude of spectroscopic applications covering almost the full electromagnetic spectrum from the UV to the terahertz.

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