substantial amount of this background radiation is removed by reflecting the heterodyne beat from a grating and partially filtering it before sending it to the photodiode.

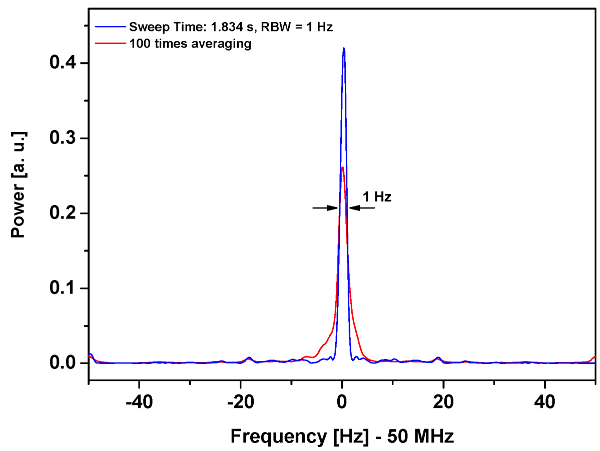

Fig. 3. Self-beat of the sum-frequency wave at 1.2 µm, between the input and the output light of the SOA. Blue: 1.8 s integration time, red: 100 averages (183 s).

In order to verify that the SOA does not add any frequency noise to the SFG wave, the amplified 1.2 µm SHG light was frequency-shifted by an AOM by 50 MHz and heterodyned in a fiber splitter/combiner with a small part of the light before the amplifier. The FWHM linewidth of this self-beat is shown in Fig. 3 and was 1 Hz, limited by the lowest bandwidth available from the spectrum analyzer (Agilent E4440A). Even at 183 s averaging time the linewidth remained at 1 Hz. This verifies that the frequency noise properties of the SOA do not add frequency noise to the sum frequency wave.

The frequency comb is based on a 250-MHz erbium-doped fiber oscillator (FC1500-250-WG, Menlo Systems). The comb is completed by three EDFA modules and a supercontinuum generation module for spectral broadening to the near-infrared (NIR) 1050 - 2100 nm spectral range. One EDFA is used as part of the carrier offset frequency (f_{ceo}) stabilization, one for two NIR output ports with combined average output power >200 mW and one for an additional port which is not used here. The last two generate a high power of >5 mW in a 3 nm wavelength window, allowing optional phase-locking of the comb to an optical reference. The long-term stable optical reference has 1-Hertz linewidth and consists of a Nd:YAG laser which is stabilized to an ultra-high-finesse ULE cavity.

Depending on the usage scenario, the comb operating mode can be selected. For investigation of wide (GHz) transitions, the comb repetition rate (f_{rep}) and the carrier offset frequency (f_{ceo}) are actively stabilized to a 10 MHz rf-signal provided by an ultrastable hydrogen maser which itself is steered to GPS. The repetition rate can be set and controlled from the comb's computer. This provides the tuning of the comb. The comb mode frequencies are given by $f_{ceo} + m\, f_{rep}$, with integer m.

If instead a narrow linewidth of the comb teeth is required, the comb can be phase-locked to an optical reference, but then it cannot be easily tuned by changing the repetition rate. The MIR- laser is then tuned by changing the beat frequency.

The amplified up-converted 1.2 µm wave and the 1.5 µm wave of the fiber laser are heterodyned in two beat note units with the respective nearest frequency comb modes. In each unit (Fig. 2), an output of the comb and a laser wave are input via respective fibers and combined in a single-mode 50% - 50% fiber splitter/combiner, allowing maximum spatial overlap. The output of the up-converted wave part is delivered into a free beam after the fiber

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combiner. Mode cleaning of the comb spectrum and wavelength filtering of the SOA radiation background is done by a grating. A $\lambda/2$ plate matches the polarization to the grating. In case of the fiber laser the comb spectrum is filtered due to a narrowband band-pass filter before coupling into the fiber combiner. The polarization of the comb spectrum is matched to the lasers' polarization by $\lambda/2$ plates also before coupling into the fiber combiners. Both heterodyne beats are detected by high-speed photodetectors. The beat notes are electrically amplified to an absolute signal level of 0 dBm. Subsequent directional couplers deliver the two beat notes Δ_1, Δ_2 to two spectrum analyzers for readout and data logging.

The beat note of the fiber laser with the comb easily reaches an SNR of 40 dB because of the small linewidth and the high power (several milliwatts) of the fiber laser beam. The beat notes are $\Delta_1 = \nu_{C\text{-}band} - (f_{ceo} + m_1 f_{rep})$ in case of the fiber laser and $\Delta_2 = \nu_{SFG} - (f_{ceo} + m_2 f_{rep})$ for the upconverted wave, respectively, where m_1 and m_2 are the mode numbers of the closest comb modes. The laser frequency stabilization loops keep Δ_1 and Δ_2 constant in time (see below).

The frequency-to-voltage detector for error signal generation in the fiber laser stabilization loop employs the Analog Devices AD8302 RF/IF gain and phase detector [45] and has a fixed locking point of Δ_1 = 44.3 MHz. For a given comb repetition rate, this frequency is initially set by fine frequency tuning of the fiber laser. An active band-pass filter before the detector filters the beat note so as to lower the noise floor.

The beat note of the upconverted 1.2 µm wave, Δ_2 is kept at 40 MHz by the QCL frequency lock. For a given comb repetition rate, this frequency is also initially set by fine frequency tuning of the QCL. For the stabilization loop it is further mixed to 64 MHz. This beat note SNR is limited to 20 - 25 dB due to the comparatively high linewidth of 1.2 MHz and the lower absolute power.

Appropriate bandpasses before and after the frequency mixer filter and isolate the beat signal, which is subsequently divided by 128 in a frequency divider, to a frequency of 500 kHz. The division of the beat note signal frequency enables a wide capturing range and a robust locking even with the low SNR of the beat note. The frequency-to-voltage detector in the QCL locking loop is based on the Analog Devices AD650 converter and has a detection range from 0 to 1 MHz, with a 0 V output for a 500 kHz signal input.

The frequencies of the QCL and of the fiber laser are each stabilized by two frequency-locking systems, adapted to the differing characteristics of the respective beat notes.

For the stabilization of the QCL, the modulation input of the current driver is used. The standard PI servo for the fiber laser acts on the piezo element inside the laser. For fixed frequency operation, the piezo control voltage stays within the allowed working range. In order to be able to scan the frequency of the fiber laser, a LabView program is used to keep the piezo control voltage within the piezo's specs by tuning the temperature setpoint of the fiber laser via its internal microcontroller.

Since long-term drifts in the beat signal frequency of the sum frequency light and the frequency comb (but not in the 1.5 µm beat) were observed during initial long-term stability measurements, a LabView-based PI control was added that adjusts the frequency of the synthesizer used to mix down the beat signal. This ensures that the beat frequency stays constant and also keeps the QCL frequency constant.

3.3 Frequency measurement and tuning

The optical frequency of the QCL is determined by the relation

$$\nu_{QCL} = (m_2 - m_1) f_{rep} \pm \Delta_2 \pm \Delta_1. \quad (1)$$

As can be seen, the carrier envelope offset frequency f_{ceo} drops out. Since all quantities on the right hand side are constant or actively kept constant in time, the QCL frequency is constant, too. The mode number difference $m_2 - m_1$, which remains constant during operation, must be known as well. m_1 and m_2 and the signs of the beat frequencies are independently

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determined from independent measurements of the wavelengths λ_{SFG} and $\lambda_{C\text{-}band}$ using a wavelength meter (Burleigh WA-1500) having an inaccuracy of less than 100 MHz. A monitor fiber output port of the fiber laser and 90:10 fiber splitter behind the 1.2 µm amplifier provide radiation of sufficient power for the wavemeter. The wavemeter is also read out by the monitoring computer and the QCL frequency is determined in the computer by applying Eq. (1).

Since f_{rep} is phase-locked to the atomic reference signal, which is also a reference for the spectrum analyzers that measure Δ_1 and Δ_2, the QCL frequency is determined in units of the atomic reference frequency, which here is 10 MHz.

The beat frequencies Δ_1, Δ_2 are measured every second by determining the weighted mean frequency of the beat spectra acquired by the two spectrum analyzers, set to average over 0.1 - 1 s, depending on the speed and step size of the repetition rate tuning, if any. This is an adequate alternative approach to the use of a frequency counter, in view of the fact that the 1.2 µm beat spectrum is rather wide. Thus, the QCL frequency is determined in real time.

Under lock, a change of the frequency comb repetition rate f_{rep} shifts the QCL frequency with a tuning coefficient given by $m_2 - m_1$. Thus, the QCL frequency can be electronically tuned by changing the frequency comb repetition rate f_{rep}. This is done by the same computer which records the two beat frequencies.

4. Results

The phase-matching temperature for the concrete SFG process used here was found to be 44 °C. However, it varies by about 2 °C depending on the power of the 1.5 µm laser, owing to absorption in the crystal at this wavelength.

The beat notes with the frequency comb when stabilized to an optical reference are shown in Fig. 4. The frequency-locked fiber laser has a linewidth of 35 kHz. The beat of the amplified 1.2 µm wave with the comb essentially represents the frequency spectrum of the QCL itself, since the linewidths of the fiber laser is relatively small. The beat linewidth is comparable to the free-running linewidth that we observed over a timescale of hundreds of ms, since the bandwidth of the employed QCL locking system is relatively low and not suitable for linewidth narrowing.

The excellent long-term stability of the comb-stabilized QCL is shown in Fig. 5. Here, the comb was stabilized to the maser only. The QCL was not tuned and the beat frequencies and the comb repetition rate value were measured once per second. Thus, the QCL frequency is computed once per second. The Allan deviation, a measure of frequency instability, is less than 10 kHz for integration times longer than 40 s. The inaccuracy of the QCL frequency is estimated as 100 kHz.

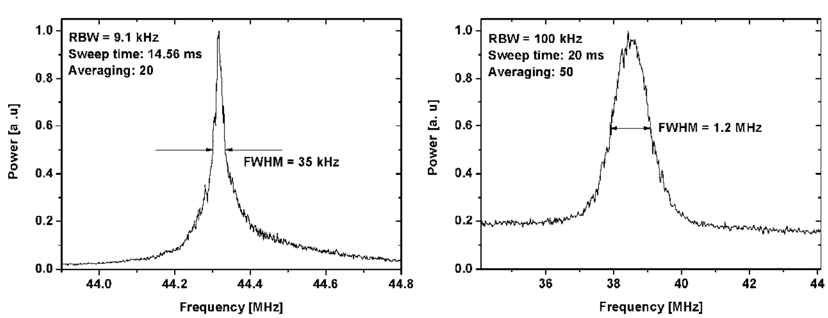

Fig. 4. Beat notes with the frequency comb which is stabilized to an optical reference. Left: Beat note of the 1.5 µm fiber laser and the frequency comb line to which it is weakly locked. Right: Beat note between the sum-frequency wave at 1.2 µm and a frequency comb line, for 1 s averaging time.

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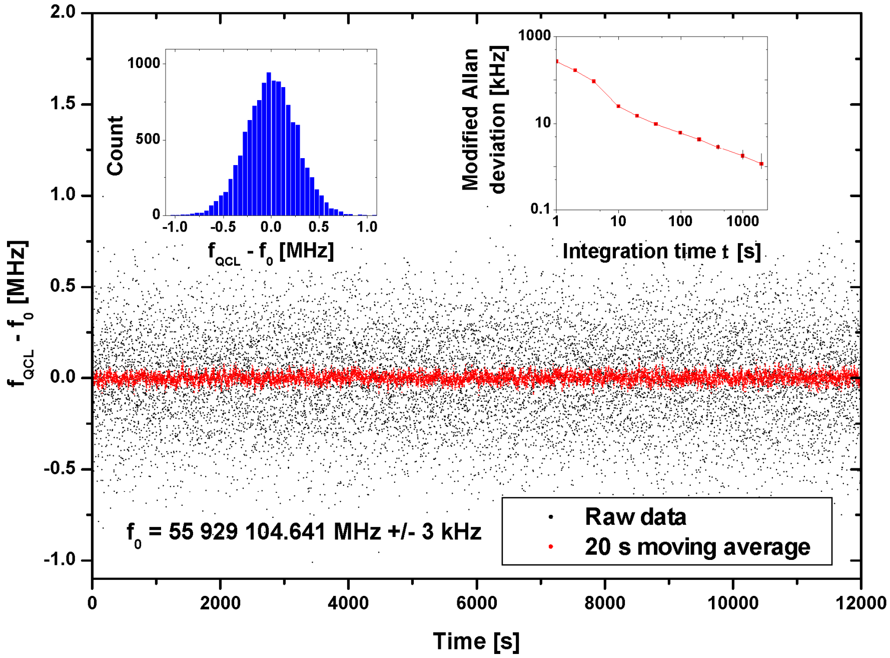

Fig. 5. Stability of the QCL frequency over three hours and histogram of frequency values and Allan deviation (insets).

The usability of the developed source for spectroscopic measurements is demonstrated by the results in Fig. 6. The QCL frequency is scanned via computer-controlled change of the comb repetition rate. This method provides a wide tuning range of at least 2.5 GHz. On purpose, we scanned the frequency very slowly, in 5000 s, to show the capability of long measurement times. The panel at the bottom of Fig. 6 shows the frequency deviations from a linear variation in time; the deviations are similar to those reported in Fig. 5 in absence of frequency scan.

In the test experiment, the QCL interrogates the P17E absorption line of the (1 1 1 0) ← (0 0 0 0) band of N_2O in a gas cell at 60 mbar. Before the QCL beam is sent through the cell, a part of the beam is split off the main beam and sent to a detector for normalization. The absorption line is shown in Fig. 6. Since the baseline of the data is not constant, a Voigt profile fit is done only using the central 600 MHz interval around the absorption peak. The fitted line center is at 55 929 343.8 ± 0.3 MHz, where the uncertainty is the 1-σ range given by the standard error of the fit (one scan). The NIST wavenumber calibration tables give 55 929 347.6 ± 2.5 MHz at a pressure of 2.7 mbar [46]). The pressure shift of the line at our pressure can be estimated to be in the range of –0.3 MHz to −5.3 MHz from pressure shifts given for other lines of the same band [47]. The difference between the two measurements therefore lies in the range of –3.4 MHz- to + 1.5 Mhz, and is consistent with zero within the error of the NIST value. However, we emphasize that our experiment was not designed for high accuracy, but only for demonstration.

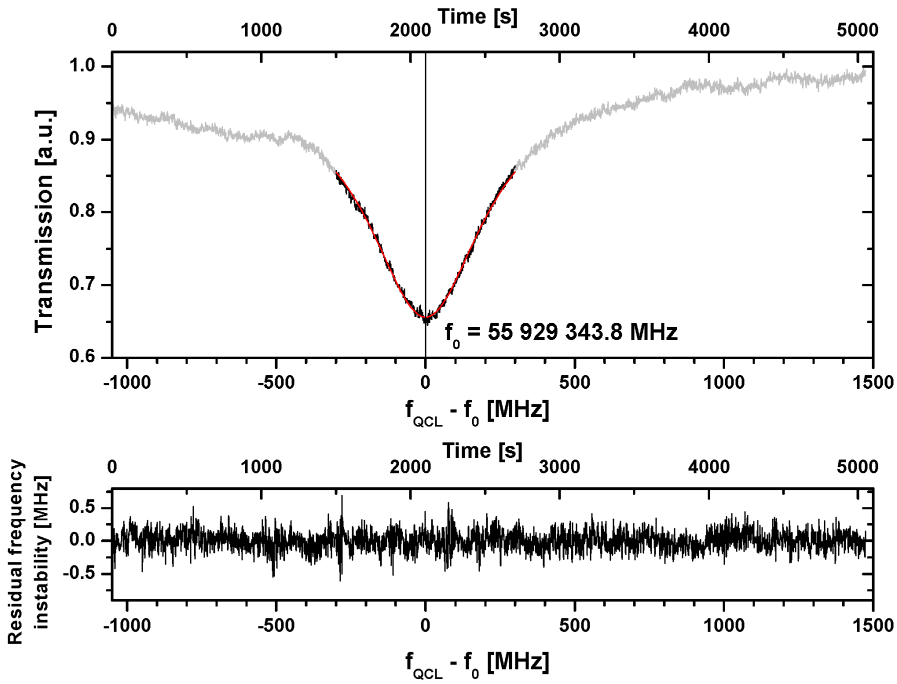

Fig. 6. Spectrometer frequency scan over a range of 2.5 GHz at a wavelength of 5.36 µm, performed by scanning the repetition rate of the frequency comb. An absorption line of N_2O was recorded simultaneously. Top: Grey line: Transmission through the gas cell versus frequency. A Voigt profile (red line) is fit to the central part of the peak (black part of the absorption peak). Bottom: measured QCL frequency minus linear fit of the frequency vs. time, showing the residual instability during the frequency scan.

6. Summary

In this paper, we have demonstrated a general method capable of providing frequency-stable radiation in the MIR range from quantum cascade lasers, with frequency absolutely known at the 100 kHz level and stable at the few kHz level. The absolute frequency is measured relative to an atomic reference.

Key element is an orientation-patterned-GaAs crystal for up-conversion of the QCL laser radiation. The crystal's properties were matched to the laser wavelength λ_{QCL} to be measured, in order to maximize the conversion efficiency of the SFG process. The coarse phase matching of the SFG process was achieved by proper choice of the QPM grating period of the OP-GaAs crystal. The precise matching was implemented by adjustment of the crystal's temperature. The fine tuning of the phase matching is also possible by tuning of the local oscillator laser's frequency.

The generated SFG radiation at 1.2 µm was amplified by using a semiconductor amplifier to a level of ca. 2 mW. We have shown that the amplification does not degrade the spectral properties. A beat with a phase-stabilized Erbium fiber frequency comb was thereby obtained with sufficient signal-to-noise-ratio. The beat permits measuring and correcting the absolute frequency fluctuations of the QCL with (currently) few kHz-level resolution.

While we have demonstrated the method for a particular MIR wavelength, and our crystal only contained two gratings, we have shown theoretically that a moderate number of OP-GaAs gratings are sufficient to cover the complete 5 – 12 µm spectral range.

The implementation of the method is robust and relatively simple. All components except the OP-GaAs crystal are standard and robust, i.e. frequency comb, atomic reference, 1.5 µm cw high-power single-frequency fiber laser, semiconductor amplifier, detectors, etc.. The

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whole system requires only short warm-up time, minimal realignments in day-to-day use and is locked within a few minutes. This is an important advantage for use of the apparatus as part of more complex experimental set-ups.

The full power of the QCL is sent through the nonlinear crystal, but, apart from reflection losses at interfaces, most of it is recovered after the crystal, and can be delivered to a spectroscopy apparatus. This means that sufficient power remains available to implement, e.g. saturation or photoacoustic spectroscopic techniques, which have higher power requirements than standard absorption spectroscopy.

In conclusion, we believe that the presented approach represents an important and flexible tool for MIR precision (i.e. comb-assisted) spectroscopy.

With its current performance and its ease of use, this type of spectrometer could be used for photoacoustic spectroscopy, multipass-cell spectroscopy, integrated cavity output spectroscopy, or Lamb-dip spectroscopy. But even as part of more sophisticated experiments, for example in the field of cold molecules [31], where the low particle temperatures require MHz-level laser linewidths, this system is suitable.

In the near future, after the development of a suitable feedback system capable of reducing further the linewidth of the QCL to the kHz level and below, high-resolution studies of molecular transitions will become possible with methods such as cavity-ring-down spectroscopy, NICE-OHMS [48], where efficient coupling into high-finesse (narrow-linewidth) optical resonators is necessary, or two-photon spectroscopy, where ultra-narrow linewidth and high power are necessary.

Acknowledgments

We thank T. Schneider and P. Dupré for helpful suggestions on the setup, and D. Iwaschko for development of electronic units. The optical reference cavity has been developed by A. Nevsky, Q. Chen, M. Cardace (Universität Düsseldorf) and U. Sterr (Physikalisch-Technische Bundesanstalt) as part of the Bundesministerium für Wirtschaft und Technologie (Germany) project 50OY1201. This work was partially funded by DFG project SCHI 431/19-1.