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Citation for the original published paper (version of record):
Richter, H., Rothbart, N., Wienold, M. et al (2024). Phase-Locking of Quantum-Cascade Lasers Operating Around 3.5 THz and 4.7 THz with a
Schottky-Diode Harmonic Mixer. IEEE Transactions on Terahertz Science and Technology, 14(3): 346-353. http://dx.doi.org/10.1109/TTHZ.2024.3385379

Phase-Locking of Quantum-Cascade Lasers Operating Around 3.5 THz and 4.7 THz with a Schottky-Diode Harmonic Mixer

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Abstract—**Quantum-cascade lasers (QCLs) are critical components for high-resolution terahertz spectroscopy, especially in heterodyne spectrometers, where they serve as local oscillators. For this purpose, QCLs with stable frequencies and narrow linewidths are essential since their spectral properties limit the spectral resolution. We demonstrate the phase-locking of QCLs around 3.5 THz and 4.7 THz in mechanical cryocoolers. These frequencies are particularly interesting for atmospheric research because they correspond to the hydroxyl radical and the neutral oxygen atom. The phase-locked loop is based on frequency mixing of the QCLs at 3.5 and 4.7 THz with the sixth and eighth harmonic, respectively, generated by an amplifier-multiplier chain operating around 600 GHz, with a Schottky diode harmonic mixer. At both frequencies we achieved a linewidth of the intermediate frequency signal of less than 1 Hz. This is about seven orders of magnitude less than the linewidth of the free running QCL.**

Index Terms—**frequency stabilization, harmonic mixers, heterodyne receivers, phase-locked loops, quantum-cascade lasers, Schottky diodes, terahertz**

I. INTRODUCTION

TERAHERTZ (THZ) quantum-cascade lasers (QCLs) are essential for high-resolution spectroscopy [1]. One application is in absorption spectroscopy, where the QCL is used as a radiation source, and the frequency-dependent absorption of its emission by, e.g., molecular transitions is detected. The other application is in remote sensing as a local oscillator (LO) in a heterodyne spectrometer for detection of atoms and molecules in astronomical objects or planetary atmospheres [2-8]. In both cases, the spectral resolution is limited by the emission linewidth of the QCL. Without any special measures, a free-running THz QCL typically has as a linewidth in the order of several MHz within a few seconds of integration time due to fluctuations of the short-term linewidth. This may increase further if the measurement time becomes longer. Fluctuations of the operating temperature, QCL driving current, and optical feedback are the foremost reasons for frequency instabilities. In contrast to the effective linewidth, an intrinsic linewidth of 90 Hz has been determined from the frequency noise power spectral density of a 2.5-THz QCL [9].

To realize a very narrow emission frequency, several approaches have been developed for the frequency stabilization of a QCL. A straightforward way is passive stabilization based on thermal and electrical bias control. This scheme relies on the fact that the QCL frequency changes with the temperature of the active medium (typically in the order of a few 100 MHz/K) and with the driving current of the QCL (typically a few MHz/mA). Keeping the temperature and current stable at the level of 1 mK and 1 µA yields a frequency-stable operation of the QCL. This scheme has been implemented in the heterodyne spectrometers GREAT and upGREAT on board of SOFIA [10, 11].

Active stabilization schemes are based on frequency references. One approach utilizes a molecular transition frequency as a reference. This approach was realized by locking the QCL frequency to an absorption line of methanol at 2.55 THz [12] and at 3.5 THz [13], respectively. The linewidth can be further improved by locking to the Lamb-dip of a molecular transition [14, 15]. With this approach, frequency stability in the order of a few hundred kHz becomes feasible.

Manuscript received XXX; revised XXX; accepted. Date of publication, XXX. This work was financed in part by the European Space Agency (ESA) under the contract No. 4000125911/18/NL/AF ”Frequency stabilisation of a Quantum Cascade Laser for Supra-THz applications” and in part by the Swedish National Space Board under the contract No. 170/17 ”THz Schottky diode mixers for high-resolution FIR spectroscopy”. *(Corresponding author: Nick Rothbart).*

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However, the gas cell is a rather challenging device for several applications, in particular for space-borne instruments. Furthermore, this stabilization scheme is limited to frequencies of molecular absorption lines and does not allow for continuous frequency tunability.

A metrology-grade THz frequency comb can be used as a stable reference for the QCL frequency. A frequency accuracy of about 1 kHz has been demonstrated for a 3-THz QCL [16]. This is limited by the accuracy of the frequency comb. The measured linewidth of the QCL is about 10 Hz. However, frequency combs for THz frequencies are still rather large devices, which limits applicability and implementation outside a laboratory environment.

The most promising approach to frequency stabilization of a QCL is using a microwave oscillator as a reference. The output signal of the oscillator is amplified and multiplied to obtain a frequency close to the QCL's frequency. Since microwave oscillators are available with linewidths below 1 Hz and frequency multiplication increases both, the linewidth and the phase noise only by 20 log(n) (n: multiplication factor), this approach provides a highly stable and precise reference frequency. A further advantage is, that the frequency can be tuned to arbitrary values simply by changing the reference frequency. Frequency and phase-locking have been demonstrated using a Schottky diode mixer [17-20], a superlattice mixer [21, 22], or a hot electron bolometer [23]. So far, the highest frequency at which phase locking has been achieved with room-temperature mixers is 3.4 THz as realized with a Schottky-diode mixer [20] and with a superlattice mixer [22].

At two higher frequencies, namely at 3.5 THz and 4.7 THz, the phase-locking of QCLs is particularly important. At 3.5 THz, the hydroxyl radical (OH) has a rotational transition, and at 4.7 THz, atomic oxygen (O) has its fine-structure ground-state transition. Both species are highly relevant in atmospheric science: OH plays a vital role in atmospheric chemistry as it is very reactive [24]. O is crucial for the energy balance of the mesosphere and lower thermosphere because it contributes to radiative cooling and is involved in exothermic chemical reactions [4, 25, 26]. In this paper, we report on phase-locking of 3.5- and 4.7-THz QCLs using a room-temperature Schottky diode harmonic mixer.

II. QUANTUM-CASCADE LASERS

The phase locking is realized for tall-barrier GaAs/Al$_x$Ga$_{1-x}$As QCLs that are based on a hybrid design combining a bound-to-continuum lasing transition with a transition resonant to the energy of the longitudinal optical phonon for efficient population of the upper laser level. Tallest barriers are achieved by employing nominally binary AlAs barriers with an effective Al content of up to $x=0.6$ [27]. The tall barriers lead to reduced electrical pump powers and consequently minimizes the heat load on the cryocooler [28, 29]. These powerful QCLs allow for the implementation of single-plasmon waveguides and straightforward Fabry-Pérot resonators, which is advantageous for many practical applications. Single-mode emission is often achieved by using short cavities without any grating structures. Based on this approach, operation in miniaturized cryocoolers was demonstrated [30].

The QCLs for 3.5 THz and 4.7 THz were manufactured at Paul-Drude-Institut für Festkörperelektronik (PDI). In total, four QCLs (two around 3.5 THz and two around 4.7 THz) were investigated (Table 1). The active regions of the QCLs are based on GaAs/AlAs (QCLs 1 – 3) or GaAs/Al$_{0.25}$Ga$_{0.75}$As heterostructures (QCL 4). The QCLs were grown using molecular beam epitaxy on semi-insulating GaAs wafers. The active regions with a doping concentration of up to 2×10^{17} cm^{-3} consist of 78 and 88 periods for 3.4/3.5 THz and 4.7/4.8 THz, respectively, with eight quantum wells in each period and a total thickness of about 11 µm for both frequencies.

TABLE 1
OVERVIEW OF THE QCLS USED IN THE EXPERIMENTS.

QCL	Frequency (THz)	Cryocooler	Resonator	Operating Point	Frequency Tuning at Operating Point
1	3.44	AIM	120 × 826 µm² Fabry-Pérot	389 mA / 54 K	~ -24 MHz/mA ~-114 MHz/K
2	3.46	Sumitomo	120 × 795 µm² Fabry-Pérot	350 mA / 20 K	~-60 MHz/mA ~-17 MHz/K
3	4.81	AIM	120 × 1019 µm² Fabry-Pérot	317 mA / 54 K	~-34 MHz/mA ~-201 MHz/K
4	4.76	Sumitomo	80 × 873 µm² DFB grating	550 mA / 20 K	~-46 MHz/mA ~-55 MHz/K

In the case of QCLs 1 – 3 (3.4, 3.5, and 4.8 THz), short Fabry-Pérot cavities ensure single-mode emission. QCL 4 (4.7 THz) has first-order lateral distributed feedback (DFB) gratings to achieve single-mode operation. The details of the investigated QCLs are given in Table 1. The operating points are the current and temperature settings used for the phase locking.

The QCLs are soldered to a copper submount, which in turn is screwed to the cold finger of a mechanical cryocooler. Two cryocoolers were used: Either a Gifford-McMahon cooler from Sumitomo (SRDK-408D) with a cooling capacity of 1 W at 4.2 K, or an AIM SL400 Stirling cooler with a cooling capacity of 1 W at 40 K. The latter one is very compact with a volume of about 300 × 120 × 140 mm^3 and a mass of 3.9 kg, and requires little electrical input power (less than 130 W) [30]. The temperature of the cold fingers of both coolers is measured with resistive temperature sensors (Cernox 1050-AA-1.5L, Lakeshore) and read out by a temperature control unit (CryoCon Model 24, Cryogenic Control Systems Inc.). For high stability of the QCL emission frequency and output power, the heat sink temperature is stabilized by a heater and a proportional-integral-differential (PID) loop to a value of ±1 mK. Both, temperature sensor and heater are mounted closely to the QCL. For laser operation, the QCL is driven by a low-noise dc current source which is combined with the phase-locked loop (PLL) electronics (ppqSense QubeCL15-P or Toptica mFalc 110).

A. Output power and frequency

For initial frequency characterization, the QCL emission frequencies were measured by a Fourier-transform spectrometer (FTS). As an example, the spectrum of QCL 4 is

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shown in Fig. 1(a), revealing single mode emission. Note that the linewidth is determined by the spectral resolution of the FTS. High-resolution molecular absorption spectroscopy was employed for a more detailed frequency characterization of the QCLs. For this measurement, all QCLs were mounted in the Sumitomo cryocooler. A polymethylpentene (TPX) lens in front of the cryocooler collimates the beam, and an absorption cell is placed between the lens and a calibrated Ge:Ga photoconductive detector. The absorption cell is filled with methanol (CH_3OH) at a pressure of 100 Pa. To obtain spectral information, the QCL current and temperature are varied in continuous-wave operation, which results in a shift of the emission frequency. As an example, the power map as a function of current and temperature of the 4.7-THz QCL in the Sumitomo cooler (QCL 4) is displayed in Fig. 1(b). The map was referenced to an absolute power measurement with a Thomas Keating (TK) power meter in front of the vacuum window of the cryocooler. The currents from 400 mA to 600 mA correspond to current densities of 573 A/cm² to 859 A/cm².

The faint narrow lines in the map are due to absorption by the CH_3OH gas. This absorption spectrum is exploited to determine the absolute emission frequency of the QCL by comparing the measured fingerprint-like absorption spectrum with the calculated transmission spectrum using the Jet Propulsion Laboratory (JPL) database [31]. A cross-cut through the current-temperature map of the 4.7-THz QCL at a temperature of 20 K and between 520 mA and 600 mA with the assigned frequency scale is shown in Fig. 1(c). The grey lines indicate JPL database entries with line intensities larger than 5×10^{-23} $cm^{-1}/(molecule/cm^2)$. The QCL frequency depends on the driving current with a negative tuning coefficient of a few 10 MHz/mA. Generally, the higher the operating temperature, the smaller is the tuning with current. The frequency tuning with temperature is also negative but with a larger variation between 6 MHz/K and 110 MHz/K. The minor deviations between the measured and the calculated frequencies indicate that the frequency tuning is not entirely linear with increasing driving current. For very high driving currents or very low temperatures, forming electric field domains in the active region leads to a significant deviation from a linear tuning behavior. Frequency calibration and the determination of the tuning coefficients were carried out the same way for all QCLs. The tuning coefficients at the operating points of the PLL are given in Table 1.

B. Beam profiles

The inset of Fig. 1(b) shows the beam profile of the 4.7-THz QCL in the Sumitomo cooler measured behind a TPX lens with a focal length of 75 mm (lens diameter: 46.4 mm) at the same distance where the aperture of the harmonic-mixer horn antenna is placed for the PLL (see section III). These profiles have been measured with a microbolometer camera for all QCLs at the same position. They are almost Gaussian-shaped, except for some small side lobes with an intensity of less than 3% relative to the maximum beam intensity. The side lobes are caused by diffraction at the TPX lens. The square indicates the aperture size of the diagonal horn antenna of the harmonic mixer. Both, QCL beam waist (230 µm) and aperture (384 µm, which corresponds to a beam waist of 165 µm) fit very well and we estimate a coupling efficiency of 75% according to Gaussian beam coupling theory [32]. The estimation is based on the optimum coupling efficiency for a diagonal horn (84%) and the mismatch between the two beams. The beam profiles of the other QCLs are similar, with somewhat lower side lobes. The estimated coupling efficiencies vary between 73% and 84% (see Table 2).

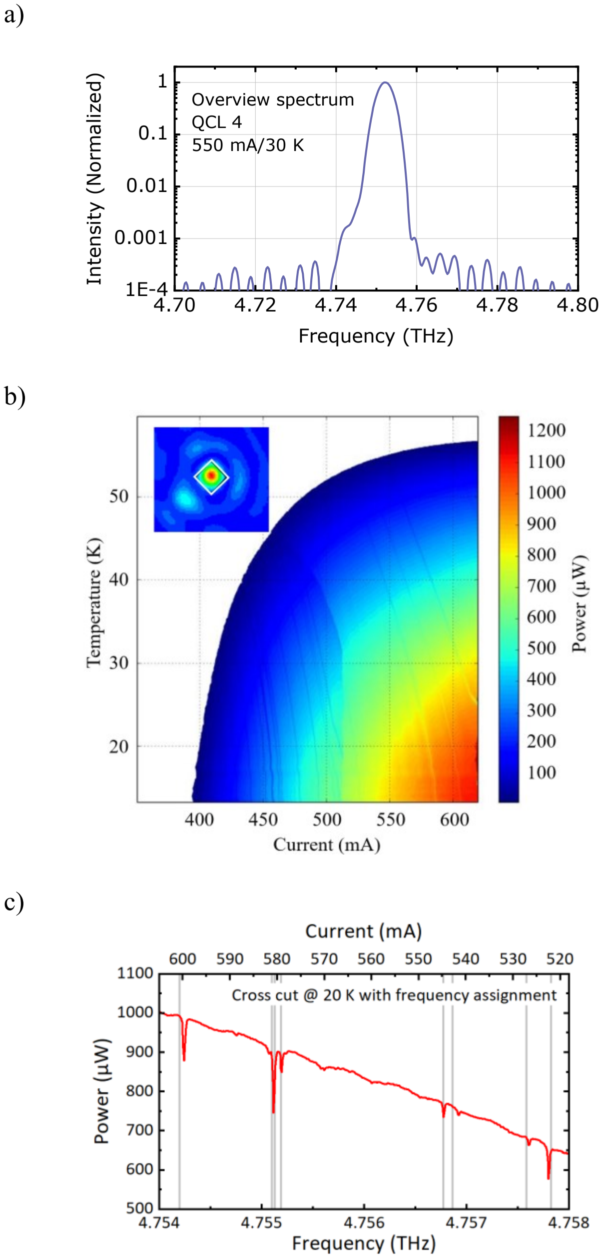

Fig. 1. (a) Overview spectrum measured with a Fourier-transform spectrometer and (b) current-temperature map of the 4.7-THz QCL in the Sumitomo cooler (QCL 4). From absorption lines of gaseous methanol (cf. narrow lines in the map), tuning coefficients of the QCL can be derived. The inset shows the beam profile at the focal point. (c) Cross-cut of the above map at 20 K with distinct absorption lines and the resulting frequency assignment (cf. lower scale).

III. PHASE-LOCKED LOOP

A scheme of the QCL stabilization with PLL is shown in Fig. 2. It is based on a Schottky diode harmonic mixer and a multiplier source, which pumps the harmonic mixer. The output frequency is tunable from about 570 GHz to 610 GHz with more than 3 mW of output power. The Schottky diode harmonic mixer generates the sixth and eighth harmonic for phase-locking of the 3.4-/3.5-THz QCLs and the 4.7-/4.8-THz QCLs, respectively. In the following sections, we will describe the main components of the PLL.

A. Multiplier source

The reference signal originates from a frequency synthesizer (Rohde & Schwarz SMA 100B) with very low phase noise (~ -40 dBc/Hz @ 1 Hz offset). It is fed into a Millitech ×8 E-band active multiplier chain (AMC-12-RNHB1). This is followed by a high-power isolator (HMI12-387-69.5-5.0 from HMI), cascaded multiplier stages, which consist of two internally power-combined doubler modules from E-band to ~300 GHz, and a single-monolithic-microwave-integrated-circuit (MMIC) varactor doubler to ~600 GHz. The multiplier MMICs have about 20% relative bandwidth. The first stage doubler was designed for optimum performance at 150-250 mW input power level with a typical conversion efficiency of 30-40%. The peak efficiency of the complete ×2 ×2 ×2 system is about 2%, with about 40%, 30% and 20% peak efficiencies for the respective doubler stages starting from the low-frequency side. The overall performance is mainly limited in frequency by the available output power from the AMC module. The output power at 600 GHz can be controlled by either varying the multiplier bias voltages or changing the active multiplier's input drive power.

B. Harmonic mixer

The Schottky diode harmonic mixer is designed to operate at the sixth harmonic of a ~600 GHz input signal [33]. It is based on a single-ended, planar Schottky diode integrated into an E-plane split mixer block. For coupling the radio frequency (RF) signal from the QCL into the harmonic mixer, a diagonal horn antenna with an aperture of 384 µm × 384 µm is machined into the mixer block [34]. The aperture dimensions of the diagonal horn correspond to a theoretical Gaussian beam waist of 165 µm [32]. A rectangular WM-64 waveguide guides the RF signal to the harmonic mixer. A WM-380 waveguide is machined into the block for the LO signal.

The measured conversion loss is about 59 dB at 3.5 THz; the mixer design and characterization details are described in [26]. While optimized initially for generation of the 6th harmonic at 3.4-3.5 THz, the same mixer also performs well with a conversion loss of 76 dB at the eighth harmonic, between 4.7-4.8 THz. This agrees well with ideal diode harmonic mixer simulations [35], showing about 10 dB difference between ×8 and ×6 harmonic operation. Therefore, the same harmonic mixer was used for the 3.5-THz PLL and the 4.7-THz PLL.

C. Intermediate frequency and phase-lock electronics

The intermediate-frequency (IF) signal is amplified by a 36-dB gain amplifier (Miteq AFS4 00100600-1310P-4), followed by another amplifier with 30 dB gain (Mini Circuits ZFL 500 LN+). The noise temperatures of the amplifiers are 110 K and 290 K, respectively. Two 3-dB attenuators reduce standing waves between the components. A 3-dB power splitter feeds half of the amplified IF signal into a spectrum analyzer while the other half is fed into the PLL electronics. For the QCLs 1 – 3, this is a commercially available device (QubeCL15-P) consisting mainly of a frequency divider, a phase-frequency detector (PFD), and a loop filter. It accepts an IF input up to 300 MHz with a lock bandwidth of 800 kHz. In the case of QCL 4, PLL electronics from Toptica (model mFalc 110) were used. The spectrum analyzer, the frequency synthesizer, and the PLL loop are synchronized to the same 10 MHz reference oscillator.

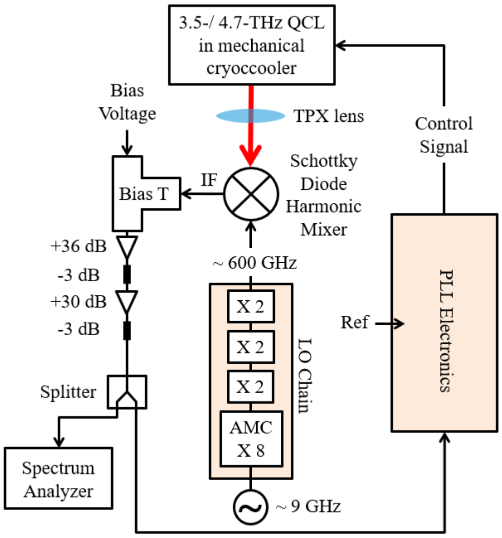

Fig. 2. Scheme of the PLL for the QCLs. The Schottky diode harmonic mixer generates the 6th harmonic (at 3.4/3.5 THz) or the 8th harmonic (4.7/4.8 THz) of the LO frequency and the IF signal of the harmonics and the QCL frequency. The PLL electronic uses the IF signal for locking the QCL frequency.

IV. PHASE-LOCKING RESULTS

A. Phase-locking at 3.4 and 3.5 THz

Phase-locking has been achieved for all QCLs listed in Table 1. At 3.4/3.5 THz, the free running linewidths are approximately 15 MHz for both QCLs as shown in Figs. 3(a) and 3(d). The spectra were acquired with a real-time signal analyzer (Keysight MXA N9020B) showing a density representing the number of times, a frequency and amplitude is hit during the capture interval. The linewidths of the free-running QCLs are determined by temperature fluctuations of the cryocoolers, mechanical vibrations, and optical feedback. In both cases, the linewidth narrows significantly when the PLL is switched on [cf. Figs. 3(b) and 3(e)]. In Figs. 3(c) and 3(f), the

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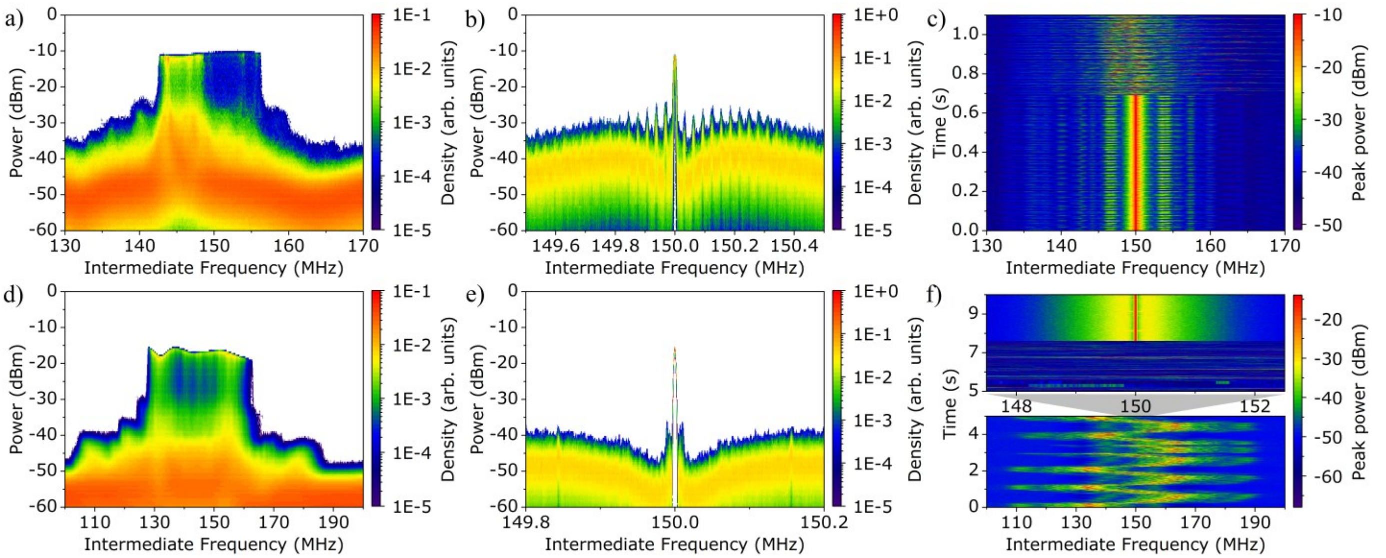

Fig. 3. Unstabilized (a, d) and stabilized (b, e) signals of the 3.4/3.5 THz QCLs mounted in the AIM cryocooler (top) and the Sumitomo cryocooler (bottom). Transitions between free-running and locked conditions are shown by waterfall plots on the right c, f).

transition from an unlocked to a locked signal is shown. In the case of the AIM cryocooler, the frequency of the unlocked signal [cf. Fig. 3(c), top] varies by about ±7.5 MHz with a period of 25 ms due to cooler vibrations. The acquisition time per spectrum of the waterfall plot was 0.1 ms. When the PLL is switched on, a narrow line with side lobes at multiples of 2 MHz appears [cf. Fig. 3(c), bottom]. The peak of the locked line is 25 dB above the peak of the first side lobe. The offset frequency of the side lobes corresponds to the bandwidth of the PLL. The situation for the QCL in the Sumitomo cryocooler is similar: Without phase-locking, a large frequency swing of about 80 MHz with a period of 1.2 s appears [cf. Fig. 3(f), bottom], which narrows when the PLL is switched on [cf. Fig. 3(f), top]. The QCL powers at the operating points of the PLLs have been measured with a TK power meter placed at the position of the horn antenna of the harmonic mixer. It was 450 µW (AIM cryocooler) and 550 µW (Sumitomo cryocooler), which was sufficient for a reliable phase-lock. Considering the estimated coupling efficiencies (cf. Table 2), the power actually coupled in is approximately 380 µW and 400 µW, respectively. Besides the coupled power the QCL, the success of the phase-lock is also strongly affected by optical feedback, vibrations of the respective coolers, PID parameters as optimized individually for each configuration, or the tuning coefficients of the QCLs. The signal-to-noise ratios (SNRs) of the locked peaks were 29 dB and 34 dB, respectively. The peak power of the side lobes is 25 dB below the maximum of the IF signal measured with a resolution bandwidth of 47.9 kHz. For comparison, phase-locking of a 3.4-THz QCL using a superlattice mixer [22] instead of a Schottky mixer also yields an IF signal with a peak power 26 dB larger than the phase noise peak with the other measurement parameters comparable (300 µW from the QCL and 100 kHz resolution bandwidth).

The fraction of the THz power which is locked in the central peak was estimated from Figs. 3(b) and 3(e) by the power in the main peak on the one hand, and the power outside of the main peak, i.e. the power in the side lobes and the noise on the other hand. It is 79 % for the QCL in the AIM cooler and 93 % for the QCL in the Sumitomo cooler.

B. Phase-locking at 4.7 and 4.8 THz

We use the same Schottky harmonic mixer for the PLL operating at 4.7/4.8 THz, since it also works very well at this high frequency. The power of the focused beams incident on the mixer was 350 µW and 360 µW, respectively (cf. Table 2). In both cases, the optics was optimized to achieve the maximum power coupled into the mixer. With beam waists of 210 µm and 230 µm, the coupling efficiencies were estimated to 79 % and 75 %, respectively. The results are displayed in Fig. 4. The free running linewidth is similar to the 3.5-THz QCLs, namely 15-20 MHz [cf. Figs. 4(a) and 4(d)], which mainly result from the cryocooler cycles (~40 Hz for the AIM cooler and ~1 Hz for the Sumitomo cooler). When activating the PLL, the linewidth reduces [cf. Figs. 4(b) and 4(e)]. In Fig. 5, spectra of the locked signal with a very high spectral resolution (479 mHz) are shown, proving the very narrow IF signal (~ 1 Hz), which is typical for the PLL. In this setting, the SNR is 55 and 65, respectively. We determined the amount of locked power to values of 93 % and 97 % from Figs. 4(b) and 4(e). The transitions from the unlocked to the locked condition are illustrated by waterfall plots in Fig. 4 (c) and 4(f). The disturbances mainly resulting from the coolers are clearly recognizable in the free-running cases. In the case of the AIM cooler [cf. Fig. 4(c)], a slow drift on top of the cooler disturbance can be seen in the free-running condition. The PLL results of all QLCs are summarized in Table 2. It should be noted, that the resulting emission frequencies can be set arbitrarily within the tuning ranges of the QCLs by changing the reference frequencies.

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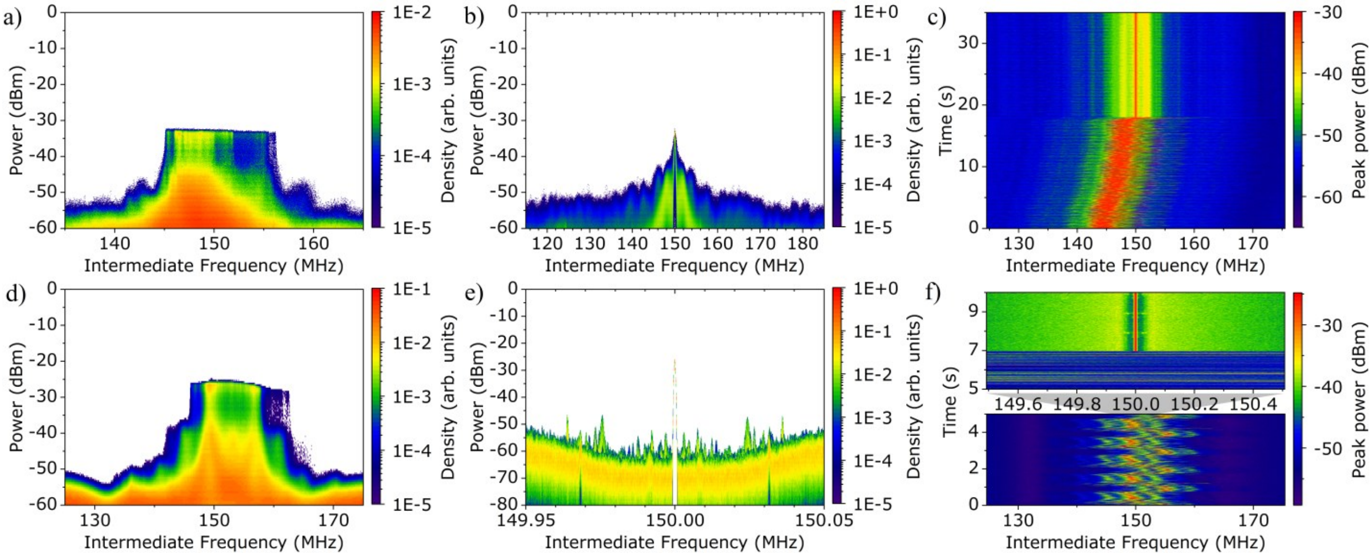

Fig.4. Unstabilized (a, d) and stabilized (b, e) signals for the 4.8 THz QCL mounted in the AIM (top) and the 4.7 THz QCL mounted in the Sumitomo cryocooler (bottom). Transitions between free-running and locked conditions are shown by waterfall plots on the right (c, f).

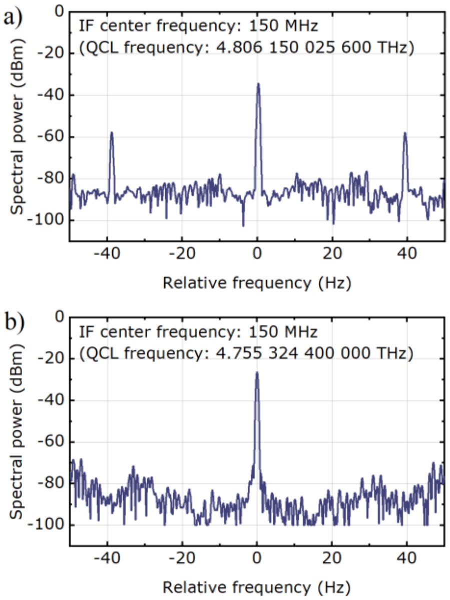

Fig.5. Highly resolved IF spectra for the 4.7/4.8 THz QCLs in the (a) AIM cooler and (b) the Sumitomo cooler.

TABLE 2
SUMMARY OF THE PLL RESULTS.

	Frequency (= LO × n + IF)	Beam Waist (µm)	Optical Coupling (%)	Power (µW)	SNR (dB)	Power in Main Peak (%)
1	**3.441 750 000 000 THz** (≈ 573.6 GHz × 6 + 150 MHz)	170	84	450	29	79
2	**3.462 221 040 000 THz** (≈ 577 GHz × 6 + 150 MHz)	240	73	550	34	93
3	**4.806 150 025 600 THz** (≈ 600.8 GHz × 8 + 150 MHz)	210	79	350	26	93
4	**4.755 324 400 000 THz** (≈ 594.4 GHz × 8 + 150 MHz)	230	75	360	39	97

V. CONCLUSION

We have successfully demonstrated phase-locking of QCLs at around 3.5 THz and 4.7 THz based on frequency mixing in a Schottky diode harmonic mixer. Narrow linewidth of the IF signal below 1 Hz were achieved with 79 % to 97 % of the power locked in the central peak. The QCLs were operated in mechanical cryocoolers, which poses an additional challenge due to their strong periodic disturbances. This proves the robustness of the PLL, which can be a significant advantage for certain application scenarios. Since the loop is referenced to a multiplier source, the QCL emission is tunable and traceable to a highly stable reference oscillator. Furthermore, stable long-term operation of the PLL is expected, since external drifts can be well compensated by the temperature control loop [10]. This offers a very large potential for high-resolution spectroscopy at terahertz frequencies. For instance, heterodyne spectrometers based on QCL local oscillators could be deployed on balloons or satellites for remote sensing of the atmosphere. Particularly interesting are the transitions of OH and O at 3.5 THz and 4.7 THz, respectively, as covered by this paper. Sensing of these transitions requires a high spectral resolution to resolve the line shape, which contains the most valuable information [4]. Consequently, phase-locked QCLs could play an important role in future missions for monitoring climate change or air pollution.

ACKNOWLEDGEMENT

While writing this manuscript, Heiko Richter passed away. He was an excellent scientist, an esteemed colleague, and a good friend. We will always remember him and dedicate this publication to his memory.

The authors would like to thank H. T. Grahn for his very valuable contribution to the development of the QCLs used for this work.

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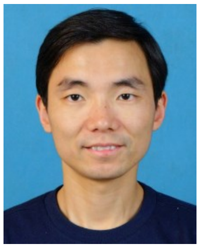

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