optica

Unambiguous real-time terahertz frequency metrology using dual 10 GHz femtosecond frequency combs

Oliver Kliebisch,[1,2,*] Dirk C. Heinecke,[1,3] Stefano Barbieri,[4,5] Giorgio Santarelli,[6] Hua Li,[4,7] Carlo Sirtori,[4] and Thomas Dekorsy[1,2]

[1]Center for Applied Photonics, University of Konstanz, 78457 Konstanz, Germany
[2]Institute of Technical Physics, German Aerospace Center, Pfaffenwaldring 38-40, 70569 Stuttgart, Germany
[3]SpaceTech GmbH, Seelbachstr. 13, 88090 Immenstaad, Germany
[4]Laboratoire Matériaux et Phénomènes Quantiques, Université Paris Diderot, 75205 Paris, France
[5]Institut d'Electronique, de Microélectronique et de Nanotechnologie, Université Lille, 59652 Villeneuve d'Ascq, France
[6]Institut d'Optique Graduate School, CNRS, Université de Bordeaux, 33400 Talence, France
[7]Key Laboratory of Terahertz Solid State Technology, Chinese Academy of Sciences, 865 Changning Road, Shanghai 200050, China
*Corresponding author: oliver.kliebisch@dlr.de

Received 20 August 2018; revised 29 September 2018; accepted 30 September 2018 (Doc. ID 341350); published 7 November 2018

Terahertz frequency metrology by radio frequency downconversion using femtosecond optical sampling relies on the harmonic factor retrieval between the terahertz frequency and the optical sampling rate. At typical femtosecond laser repetition rates, this imposes an ambiguity for frequency metrology. We report on a dual-comb sampling system for the unambiguous frequency measurement of terahertz quantum cascade lasers with hertz-level precision. Two Ti:sapphire oscillators with 10 GHz repetition rate are used for the electro-optic sampling of terahertz radiation at 2.5 THz emitted by actively mode-locked terahertz quantum cascade lasers with 9.7 GHz and 19.6 GHz repetition rates. By coherent downconversion, the emitted terahertz waveforms are measured in the radio frequency domain. The terahertz frequency comb is stabilized by employing a phase-locked loop on a radio frequency beat-note signal. A second infrared sampling comb is used to measure the absolute frequencies of the terahertz radiation. This method, which is based on the detuning of the sampling repetition rates, allows the direct retrieval of the quantum cascade laser's absolute frequency in real time without using additional optical frequency references for calibration. In order to demonstrate the feasibility of the stabilization and readout technique, a high-resolution spectroscopy measurement on gaseous methanol is presented.

https://doi.org/10.1364/OPTICA.5.001431

1. INTRODUCTION

Terahertz quantum cascade lasers (QCLs) play an important role as terahertz sources for high-resolution spectroscopy [1], due to their very narrow intrinsic linewidth [2,3], high output power, and potentially compact form factor as compared to terahertz generation using nonlinear effects or photoconductive emitters. In the terahertz spectral region, the rotational transitions of simple molecules have Doppler-limited linewidths in the range of a few 100 kHz, requiring metrology-grade tools for high-resolution spectroscopy. To achieve a subkilohertz resolution is challenging in common Fourier-transform spectrometers or far-infrared laser-based spectrometers. Using calibrated QCLs for absorption spectroscopy can provide a powerful tool to catalog the molecular transitions in the terahertz frequency band [1]. The challenge to use terahertz QCLs for frequency metrology lies in the measurement of the absolute frequency values of the QCL modes. This is possible by using coherent sampling of the terahertz waveform by a femtosecond laser [4]. In previous works [5,6], the measurement of the absolute frequency is limited by the ambiguity of the harmonic factor between the unknown frequency and the optical sampling rate. In Ref. [5], two sampling combs in the hundred megahertz range have been used, resulting in harmonic index jumps and difficulties due to the drifting repetition rate difference between the sampling combs. The experiment presented in [6] used a single stabilized 77.5 MHz repetition rate Ti:sapphire laser and a lHe cooled hot-electron bolometer as frequency downconversion element. By tuning the repetition rate, a harmonic index measurement is performed, limited by the residual QCL frequency jitter, and thus a high uncertainty on the harmonic index. Applying the frequency measurement to high-resolution spectroscopy required elaborate techniques to identify the measured absorption feature.

We present the implementation of a dual-comb sampling technique using high repetition rate femtosecond Ti:sapphire lasers.

The high repetition rate, together with the dual sampling technique, completely eliminates the frequency ambiguity. Therefore, this method allows the instantaneous and unambiguous retrieval of the QCLs' absolute frequency without using additional optical frequency references for calibration, providing an absolute frequency reference in the terahertz region.

2. DIRECT FREQUENCY READOUT

Two terahertz QCLs with different cavity lengths are used for the presented experiment. They are based on a bound-to-continuum active region design with a single plasmon waveguide [7] and are operated in the longitudinal multimode regime, with frequencies ranging from 2.4 to 2.6 THz. By injecting a radio frequency (RF) bias modulation close to the respective round-trip frequency of the QCLs, active mode-locking is achieved [8]. Compensating for losses in the RF components, typically 0 to 5 dBm of RF power have been used for injection locking. The first QCL is a 4.03 mm long device with a longitudinal mode spacing of about 9.73 GHz and a total output power of 5 mW. It was used for all presented results on dual-comb sampling and phase locking. Due to better spectral overlap with methanol absorption lines, a second QCL is used for the spectroscopy experiments presented in this paper. The second QCL has a length of 1.99 mm, resulting in 19.59 GHz longitudinal mode spacing; the delivered power is up to 15 mW.

In order to measure and stabilize the absolute frequency of the terahertz QCL, the electric field is sampled using coherent asynchronous optical sampling at a repetition rate of 10 GHz [4]. The experimental implementation is shown in Fig. 1. Two Kerr lens mode-locked femtosecond Ti:sapphire lasers with $f_{R,f_s} =$ 10 GHz and $f_{R,f_s} + \Delta f_{R,f_s}$ repetition rate [9] are used as sampling combs. The absolute repetition rate of the master oscillator is phase-locked to an RF synthesizer, while the repetition rate of the slave oscillator is stabilized relative to the first oscillator [10]. The sampling combs' repetition rate offset $\Delta f_{R,f_s}$ is chosen to be typically in the range of 100 to 1000 kHz. The repetition rate stability is $\delta f_{R,f_s} = 2$ mHz, and the repetition rate offset stability is $\delta\Delta f_{R,f_s} = 288$ μHz. The two femtosecond lasers are noncollinearly focused through the same $f = 300$ mm lens on a 2 mm thick ZnTe crystal passing through a cone-shaped bore in an off-axis parabolic mirror with an effective focal length of 25.4 mm. The terahertz beam spatially overlaps both femtosecond laser beams, while the spatial overlap among the Ti:sapphire lasers is kept to a minimum to avoid cross-correlation artifacts. The following quarter-wave plate and polarizing beam-splitter cube complete the unbalanced electro-optic detection. The unbalanced

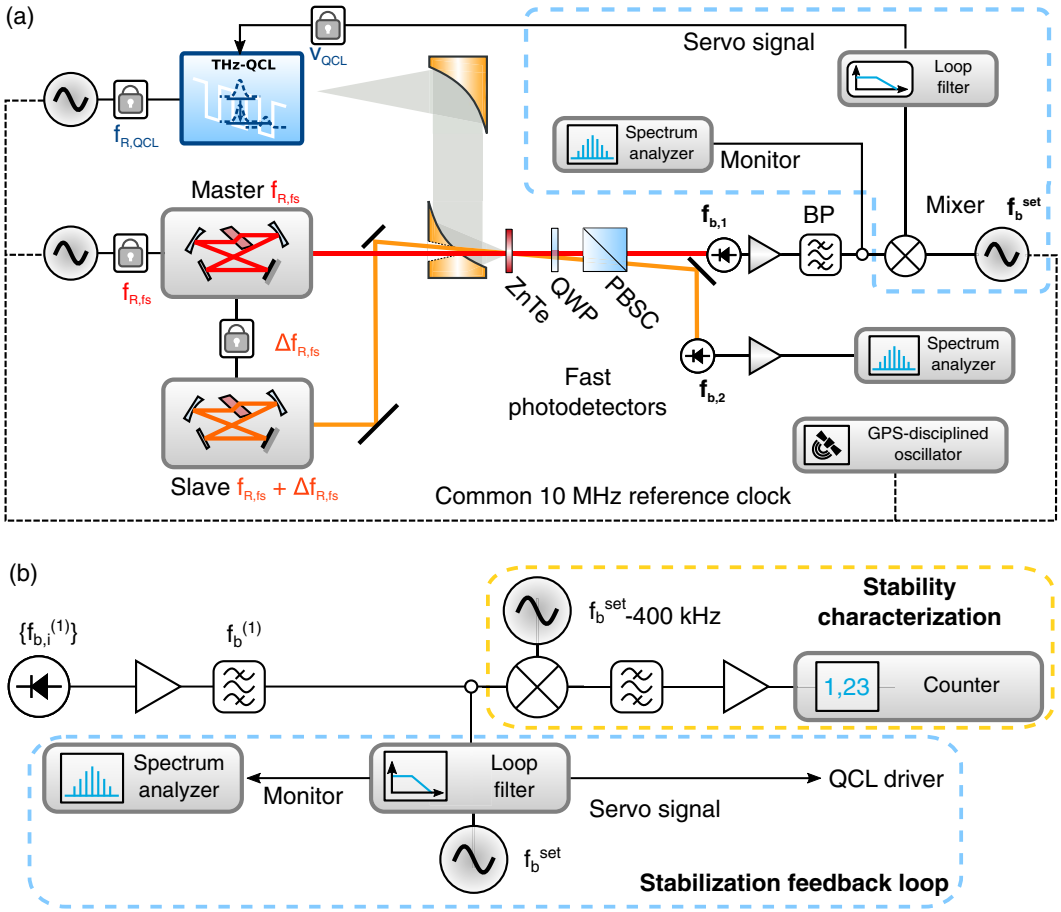

Fig. 1. (a) Schematic overview of the dual-comb sampling setup. The two femtosecond lasers have the same wavelength but are depicted in red and orange for visual clarity. The terahertz beam path is indicated in gray. The dashed black line marks the common 10 MHz reference clock shared among all synthesizers. The dashed light blue path matches the stabilization feedback loop, which is shown in more detail in Fig. 1(b). A full description is given in the text. ZnTe, 2 mm thick ZnTe crystal; QWP, quarter-wave plate; PBSC, polarizing beam-splitter cube; BP, electronic bandpass filter; (b) detailed view of the signal conditioning of the first photodiode for frequency stabilization and in-loop characterization. After the longitudinal beat-mode spectrum is amplified, and a single mode is filtered, the signal is split into a feedback loop branch and an in-loop characterization branch [not shown in Fig. 1(a)].

detection allows the use of higher probe powers without saturating the photodetectors [11]. The probe power incident on the electro-optic crystal was 300 mW for both femtosecond lasers. The intensity-modulated femtosecond lasers are detected on two separate amplified Si-photodetectors. Following the derivation in [4], the sampled waveform $E_S(t)$ at the detector of the master oscillator can be described as

$$E_S(t) = E_{\mathrm{QCL}}(t) \cdot \mathrm{III}_{f_{R,fs}}(t), \tag{1}$$

$$= \exp(i \cdot 2\pi\nu_{\mathrm{QCL}} \cdot t) f_{R,fs} \times \sum_{m=-q}^{p} E_m \exp(i[2\pi m f_{R,\mathrm{QCL}} + \phi_m])$$

$$\times \sum_{k=-\infty}^{\infty} \exp(i \cdot 2\pi k f_{R,fs}) + cc. \tag{2}$$

Here $\mathrm{III}_{f_{R,fs}}(t)$ is the Dirac comb function with a spacing of $f_{R,fs}$, and $E_{\mathrm{QCL}}(t)$ is the electric field of the terahertz QCL. The QCL field is described by a sum of $M = q + p$ planar waves with amplitude E_m and phase ϕ_m. The spectrum of the QCL is a comb with carrier frequency ν_{QCL} and repetition rate $f_{R,\mathrm{QCL}}$. It can be shown that Eq. (2) describes a downconverted RF signal. By introducing the new carrier (beat-)frequency

$$f_b = \nu_{\mathrm{QCL}} - n \cdot f_{R,fs}, \qquad n = \mathrm{int}\left(\frac{\nu_{\mathrm{QCL}}}{f_{R,fs}}\right), \tag{3}$$

and the new mode spacing

$$\Delta\tilde{f} = f_{R,\mathrm{QCL}} - r \cdot f_{R,fs}, \qquad r = \mathrm{int}\left(\frac{f_{R,\mathrm{QCL}}}{f_{R,fs}}\right), \tag{4}$$

the RF waveform can be expressed as

$$E_S(t) = \exp(i \cdot 2\pi f_b \cdot t) f_{R,fs} \times \sum_{m=-q}^{p} E_m \exp(i[2\pi m \Delta\tilde{f} \cdot t + \phi_m])$$

$$\times \mathrm{III}_{f_{R,fs}}(t) + cc. \tag{5}$$

From Eq. (3), it becomes clear that this sampling technique provides a direct link of the absolute QCL frequency to the RF domain. If the beat frequency f_b is measured and the harmonic index n is known, the QCL frequency can be measured with a precision only limited by the femtosecond lasers' repetition rate stability $n \cdot \delta f_{R,fs}$. To determine the harmonic index, the second RF beat signal is measured by sampling with the slave oscillator using a different repetition rate [12]. As both femtosecond lasers experience the same terahertz field, only the beat signal frequencies will vary due to the difference $\Delta f_{R,fs}$ in the sampling frequency. As long as the sign of the beat frequency is the same for both sampling combs (i.e., $0 < f_b \pm n \cdot \Delta f_{R,fs} < f_{R,fs}/2$), the harmonic index can be calculated from the two measured beat signal frequencies given by Eq. (3):

$$n = \frac{\Delta f_b}{\Delta f_{R,fs}}. \tag{6}$$

Assuming $\Delta f_{R,fs}$ to be stable, the error of the harmonic index can be estimated as

$$\delta n = \frac{1}{\Delta f_{R,fs}} \delta(\Delta f_b) = n \times \frac{\delta(\Delta f_b)}{\Delta f_b}. \tag{7}$$

As n is an integer number, the harmonic index can be unambiguously determined if the beat frequency difference can be measured with a precision better than $\Delta f_{R,fs}/2$. Because n is small (≈250–260) for this high-repetition rate sampling system, the required measurement precision is in the 100 kHz range compared to typical femtosecond oscillator repetition rates in the subgigahertz region ($n \gg 1000$), which would require a much higher beat frequency stability. This is expressed by the second representation of δn in Eq. (7). The harmonic index error is n times the relative error of Δf_b. Typically, $\Delta f_{R,fs}$ is chosen to fit the whole beat frequency spectrum within the range of $0 < f_b < f_{R,fs}/2$ without any index jumps, which further limits the maximum value of $\Delta f_{R,fs}$ for lower repetition rate systems.

To ensure the unambiguous frequency measurement, the terahertz frequency comb produced by the QCL is stabilized using a phase-locked loop described in Fig. 1(b). The downconverted waveform described by Eq. (5) consists of a set of beat frequencies $\{f_{b,i}^{(1)}\}$. The beat frequency spectrum is amplified, and one mode is selected to be fed into the phase detection of the feedback loop. In the loop filter electronics (described in [13]), the selected mode is mixed with an RF signal at frequency f_b^{set}, defining the desired locking frequency. After phase detection, a proportionally-integrating (PI)-filter generates a servo signal, which is fed into the modulation input of the QCL constant current driver. For characterization of the feedback loop, a monitor signal is analyzed with an electronic spectrum analyzer. Additionally, the beat-mode signal is measured using a frequency counter. In order to decrease the broadband RF noise floor, which limits the frequency counter's sensitivity, the signal is frequency-shifted to 400 kHz by mixing it with another RF synthesizer and band-limited using a narrow bandpass filter centered at 400 kHz. Phase coherence between all RF synthesizers used, the spectrum analyzers, and the counter is established by distributing a common 10 MHz reference signal of a GPS-disciplined Rb-clock.

3. DUAL-COMB SAMPLING AND FREQUENCY METROLOGY RESULTS

A typical beat-mode spectrum of the QCL at 1.3 A current and 21 K operating temperature is presented in Fig. 2(a). A GaAs photodetector with 12.5 GHz bandwidth has been used. The signal was measured with 3 kHz resolution bandwidth. The spectrum consists of 12 modes in this case. From this beat-mode spectrum, a single line at $f_b^{(1)} = 606$ MHz is selected to be fed into the frequency stabilization scheme. Figure 2(b) shows the monitor signal of the phase-locked loop in the closed-loop state as well as the derived signal for frequency counting in a small span around $f_b^{(1)}$. Two sidebands originate from the P-share of the loop filter, indicating a loop bandwidth of about 600 kHz. The signal-to-noise ratio of the locked beat signal at different resolution bandwidth is shown in Fig. 2(c). Fitting the measured values shows a slope of −9.6 dB/decade, which indicates that almost all power of the mode is locked using the phase-locked loop. The central peak of the monitor signal is isolated using the described filtering scheme and fed into a frequency counter. The frequency stability over time is shown in Fig. 2(d). The dashed lines indicate the +/− 5 times the median absolute deviation (MAD) range. With $y(i)$ as the normalized frequency samples, the following MAD definition is used [14]:

$$\mathrm{MAD} := \frac{\mathrm{median}|y(i) - \mathrm{median}[y(i)]|}{K_{\mathrm{MAD}}}. \tag{8}$$

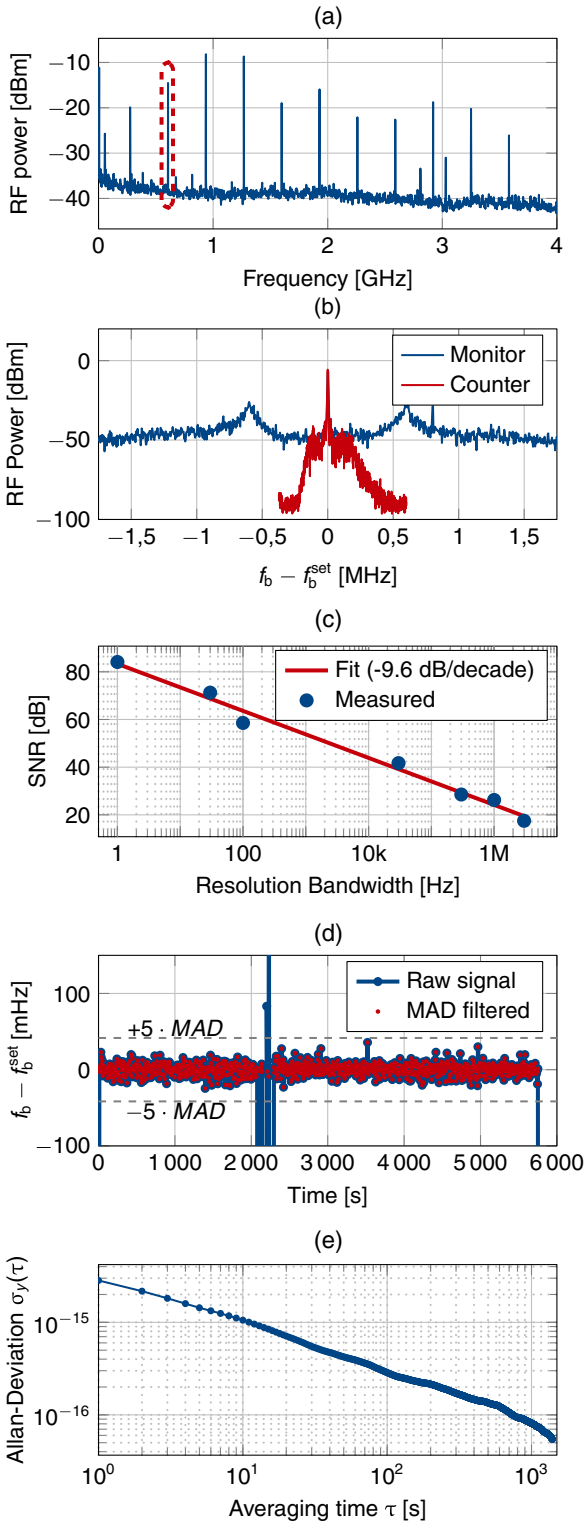

Fig. 2. (a) Longitudinal beat-mode spectrum sampled by the master femtosecond laser. The dashed red line indicates the selected beat mode for frequency stabilization and characterization. (b) Detailed spectrum of the phase-locked loop monitor signal and the 400 kHz filtered counting signal. Both spectra have been acquired using 3 kHz resolution bandwidth. (c) SNR of the locked beat signal versus resolution bandwidth of the spectrum analyzer; (d) plot of the frequency deviation from the desired beat-mode frequency f_b^{set} over time. The dashed lines indicate the $5 \cdot \mathrm{MAD}$ filter criterion for outlier removal. (e) Logarithmic $\sigma - \tau$ plot of the overlapping Allan deviation.

The scaling constant is set to $K_{\mathrm{MAD}} = \sqrt{2}\,\mathrm{erf}^{-1}(1/2)$, which results in the $5 \cdot \mathrm{MAD}$ being equivalent to a $5 \cdot \sigma$ threshold for normally distributed samples. Using the $5 \cdot \mathrm{MAD}$ criterion for outlier removal, the overlapping Allan deviation in Fig. 2(e) is calculated. The values have been normalized to the QCL central frequency of $\nu_{\mathrm{QCL}} = 2.52$ THz. The relative frequency stability of the beat signal is 2.8×10^{-15} at $\tau = \tau_0 = 1$ s, and a dead-time-limited $\propto \tau^{-1/2}$ behavior is observed. As this measurement scheme characterizes the in-loop stability, it follows from Eq. (3) that the frequency stability of ν_{QCL} is only limited by the absolute repetition rate stability amplified by the harmonic index $n \cdot \delta f_{R,f_s}$. As the harmonic index is $\approx n = 250$ for the 2.52 THz mode, the absolute frequency stability of the QCL is on the order of 500 mHz using this 10 GHz femtosecond laser for electro-optic sampling.

The precise determination of the absolute frequency of the QCL requires the exact knowledge of the harmonic index, which is *a priori* unknown when using only one femtosecond sampling comb. An exemplary measurement of the beat frequency spectrum using two femtosecond combs is shown in Fig. 3. Both beat modes originate from the same longitudinal mode of the QCL, but due to a difference of $\Delta f_{R,f_s} = 500$ kHz in the sampling combs, mode spacing shows a beat-mode frequency separation of $n \cdot \Delta f_{R,f_s}$. Applying Eq. (6), a harmonic index of $n = 256$ is determined for this particular mode. The terahertz frequency and all parameters are summarized in Table 1. It should be highlighted that the beat frequencies have been measured using the

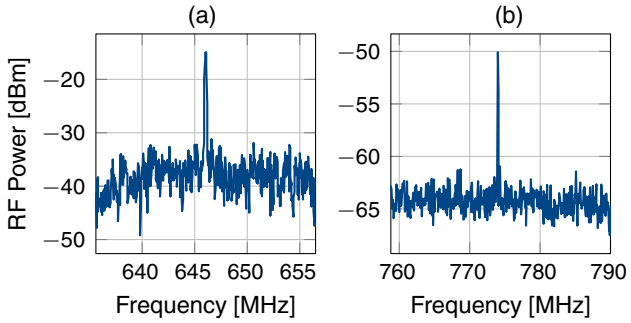

Fig. 3. Electronic spectrum of the simultaneously measured beat-mode signals as sampled by the master laser (a) at 10 kHz resolution bandwidth and the slave laser (b) at 3 kHz, which stem from the same longitudinal mode of the QCL.

Table 1. Set of Parameters for Determining the Absolute Frequency from the Beat Signals Shown in Fig. 1

Parameter	Set Value
$f_{R,f_s}^{(1)}$	9.98995000 GHz
$\Delta f_{R,f_s}$	500.00 kHz
Parameter	**Meas. Value**
$f_b^{(1)}$	646.00000 MHz
$f_b^{(2)}$	774.00000 MHz
Parameter	**Calc. Value**
Δf_b	128.00000 MHz
n	256
ν_{QCL}	2.55678120000 THz

internal frequency counters of the electronic spectrum analyzers used. The internal frequency counters of the spectrum analyzers were set to 1 Hz resolution. The frequency error of the counter is on the hertz level and is therefore negligible. The frequency error of the frequencies stabilized to synthesizers is of the same level. Assuming an accuracy of the set and measured frequencies of 10 Hz, the uncertainty of the harmonic index is $+/-0.00051$, or better than $5 \cdot 10^{-6}$. As the harmonic index is an integer number, this results in the unambiguous and exact retrieval of the index. The high repetition rate of the sampling combs allows the use of larger repetition rate offsets, resulting in relaxed uncertainty constraints for determining the harmonic index and thus the absolute QCL frequency with hertz-level precision.

4. METHANOL SPECTROSCOPY SETUP

As an application and demonstration of the outlined frequency readout technique, a high-resolution absorption spectroscopy experiment in gaseous methanol has been carried out. The extension of the frequency metrology setup for spectroscopy is shown in Fig. 4. The collimated beam diameter is approximately 30 mm. The terahertz beam is guided onto a 55/45 silicon beam splitter, which directs a portion of the terahertz power through a 23 cm long gas cell filled with gaseous methanol. The cell windows made of polytetrafluoroethylene (PTFE) are aligned at an angle of 5°. The wedge angle of the windows is 0.5°. The beam is subsequently focused onto a pyroelectric detector using a $f = 50$ mm TPX lens. This involves chopping of the terahertz beam using a mechanical chopper wheel at $f_{\mathrm{mod}} = 20$ Hz modulation frequency, which is generated by the lock-in amplifier. The signal of the pyroelectric detector is amplified using a transimpedance amplifier with 1×10^8 V/A transimpedance gain and demodulated by the lock-in amplifier.

The spectroscopy experiment has been carried out using the second QCL with 19.59 GHz mode spacing, as it showed better spectral overlap with stronger methanol absorption lines. The Fourier-transform infrared (FTIR) spectrum of the QCL is shown in Fig. 5. The specified FTIR resolution is 7.5 GHz. The absorption experiment has been conducted at about 1.35 A drive current and temperatures between 20 and 27 K. It should be noted that the spectral power distribution is highly sensitive to external cavity feedback from the ZnTe crystal. The feedback is minimized by tilting the ZnTe crystal with respect to the parabolic mirrors, but any movement of the ZnTe crystal will change the feedback to the QCL and therefore its spectral properties.

The absolute frequency measurement scheme that has been characterized above is applied in exactly the same manner. This ensures the precise tracking of longitudinal mode frequencies

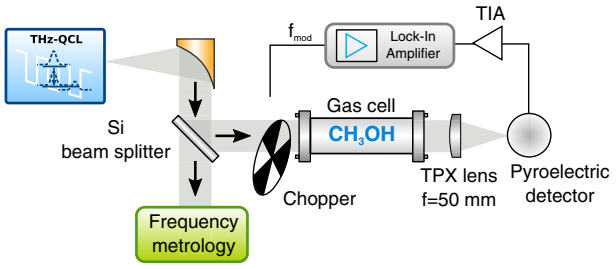

Fig. 4. Schematic of the extended setup for absorption spectroscopy. The part labeled frequency metrology is identical to Fig. 1. TPX, polymethylpentene; TIA, transimpedance amplifier.

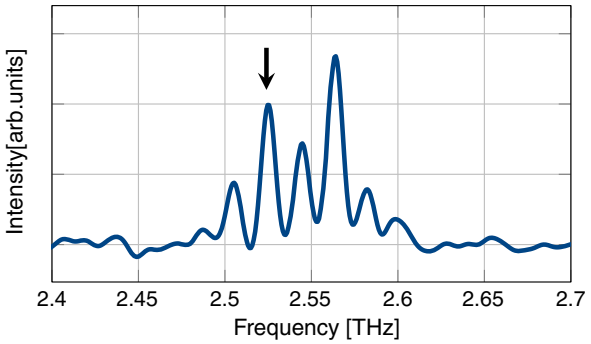

Fig. 5. FTIR spectrum of the QCL with 19.6 GHz longitudinal mode spacing at 21 K temperature and 1.4 A drive current. The QCL mode, which is tuned to a methanol absorption line, is indicated by the black arrow.

of the QCL while tuning to a methanol absorption feature. The addressed absorption line is an E2 transition with a line center at about 2.524 THz, which is listed in the Jet Propulsion Laboratory (JPL) spectroscopic database [15] and has been experimentally measured in other works, e.g., [1,16].

In multimode or mode-locked operation, the distribution of the QCL power on all longitudinal modes changes rapidly when the drive current of the QCL is changed. Therefore, the temperature and current setpoints have been chosen to allow temperature-tuning of the QCL frequency over a range of 50–100 MHz without substantial power fluctuations in the longitudinal mode close to the methanol absorption line. The frequency has been tuned by changing the local oscillator frequency in 100 to 500 kHz steps. A software control loop ensures that the phase-locked loop is closed and modifies the QCL temperature to shift the QCL into the locking range of the phase-locked loop (PLL) if the lock has been lost.

5. RESULTS ON HIGH-RESOLUTION SPECTROSCOPY

The measured absorption is shown for two different nominal pressure levels in Fig. 6. The nominal pressure has been controlled

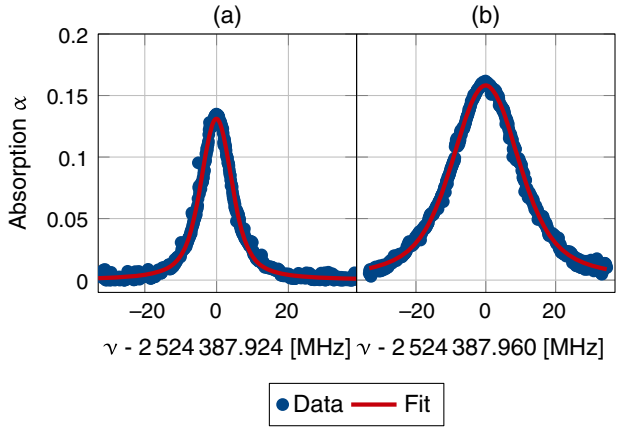

Fig. 6. Absorption spectra of methanol for the observed E2 transition at (a) 0.4 mbar and (b) 0.85 mbar nominal pressure, respectively. The fitted Voigt profile is shown in red.

Table 2. Measured Absorption Line Is an E2 Transition with Rotational Quantum Number $J'' = 26 \rightarrow J' = 25$, Projection Quantum Number $K'' = -8 \rightarrow K' = -7$, and Torsion Quantum Number $v_t'' = 0 \rightarrow v_t' = 0$

	This Work				
	$p = 0.4$ mbar	$p = 0.85$ mbar	$p = 0$ (extrapolated)	JPL Database [15]	Pearson [1]
Intensity S_{ij} [cm^{-1}/(molecule/cm^2)]	–	–	–	8.5×10^{-22}	–
Line center ν [THz]	2.524387924	2.524387960	2.524387892	2.524387881	2.524387914
Uncertainty $\delta\nu$ [kHz]	17	23	53	39	100
Line width [MHz]	10.3	23.1	–	–	–

using a vacuum pressure gauge (Instrutech Stinger). The raw voltage recorded using the lock-in amplifier has been corrected by a linear background stemming from the power change of the longitudinal QCL mode when tuning the QCL temperature. Due to the pressure shift, the line center is different between the two measurements [see Fig. 6(a) and 6(b)]. The measured data is fitted using a Voigt model, though the line shape is still mostly determined by pressure broadening in this parameter regime. The determined fit parameters, their uncertainties, and the pressure shift corrected value are listed in Table 2. The line center uncertainty for both pressure values is on the order of 20 kHz; suggesting an absolute precision of 10^{-8}. Figure 7 shows a plot of the observed line centers at different pressure levels for this work in comparison to [1] and [15]. A simple linear extrapolation of our observations to $p = 0$ results in the pressure shift corrected value of 2.524387892 THz with an uncertainty of 53 kHz. This result is in close agreement and within the uncertainty bounds of the JPL database [15] and Pearson's work [1]. Adding to the frequency measurement precision of at least 10^{-8}, the absolute frequency accuracy is also on the order 10^{-8} if the JPL database is taken as the frequency reference source.

This measurement puts a lower limit on the achievable absolute precision using the stabilized QCL, as the determined absolute accuracy is still much higher compared to the relative precision, which is on the order of 10^{-12}. By improving the experimental pressure control and measurement within the gas cell, the pressure shift parameter could be determined using this spectroscopy technique. This allows one to determine the line center more precisely. As reported in [6], other effects like Zeeman shift, ac-Stark shift or blackbody shift could be observed on the sub-1 kHz precision regime.

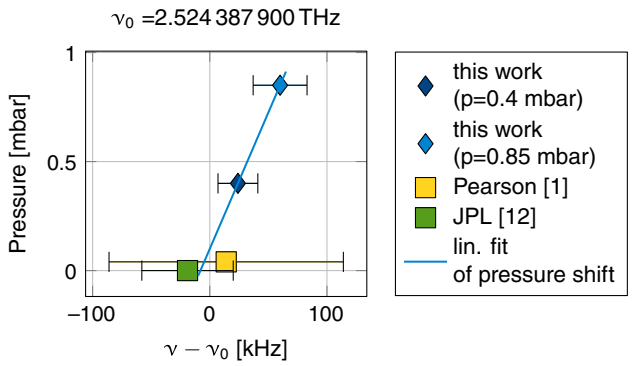

Fig. 7. Plot of the pressure-induced line center shift observed in this work in comparison to other works on this particular E2 transition.

6. CONCLUSION

We have presented the dual-comb sampling of an actively mode-locked terahertz QCL using two near-infrared frequency combs with 10 GHz repetition rate. This technique allows for the unambiguous absolute measurement of the frequencies emitted by the QCL with hertz-level precision. In this way, the QCL can be traced to a microwave frequency standard and used for metrology-grade high-precision spectroscopy in the terahertz spectral region [6,17]. The stability is ultimately limited by the multiplication of two factors. The first one is the ratio between the sampled terahertz radiation frequency and the sampling combs' repetition rate (harmonic index); the second one is the stability of the absolute repetition rate lock of the master sampling comb. Apart from the cryogenic cooling of the QCL, this method relies on room-temperature detection methods and does not require hot electron bolometers for beat-note generation. In future prospect, the high power of the QCL in the milliwatt range should allow for Doppler-free saturated absorption spectroscopy.

Funding. Carl-Zeiss-Stiftung; Deutsche Forschungsgemeinschaft (DFG).

Acknowledgment. Oliver Kliebisch thanks the Carl-Zeiss-Stiftung (Stuttgart, Germany) for financial support. We thank Patricia Scheel and Arne Budweg for regular support with the operation of the cryogenic equipment.

REFERENCES

1. J. C. Pearson, B. J. Drouin, S. Yu, and H. Gupta, "Microwave spectroscopy of methanol between 2.48 and 2.77 THz," J. Opt. Soc. Am. B **28**, 2549–2577 (2011).
2. M. Ravaro, S. Barbieri, G. Santarelli, V. Jagtap, C. Manquest, C. Sirtori, S. P. Khanna, and E. H. Linfield, "Measurement of the intrinsic linewidth of terahertz quantum cascade lasers using a near-infrared frequency comb," Opt. Express **20**, 25654–25661 (2012).
3. M. S. Vitiello, L. Consolino, S. Bartalini, A. Taschin, A. Tredicucci, M. Inguscio, and P. De Natale, "Quantum-limited frequency fluctuations in a terahertz laser," Nat. Photonics **6**, 525–528 (2012).
4. S. Barbieri, M. Ravaro, P. Gellie, G. Santarelli, C. Manquest, C. Sirtori, S. P. Khanna, E. H. Linfield, and A. G. Davies, "Coherent sampling of active mode-locked terahertz quantum cascade lasers and frequency synthesis," Nat. Photonics **5**, 306–313 (2011).
5. M. Ravaro, V. Jagtap, C. Manquest, P. Gellie, G. Santarelli, C. Sirtori, S. P. Khanna, E. H. Linfield, and S. Barbieri, "Spectral properties of THz quantum-cascade lasers: frequency noise, phase-locking and absolute frequency measurement," J. Infrared Millimeter Waves **34**, 342–356 (2013).
6. S. Bartalini, L. Consolino, P. Cancio, P. De Natale, P. Bartolini, A. Taschin, M. De Pas, H. Beere, D. Ritchie, M. S. Vitiello, and R. Torre, "Frequency-comb-assisted terahertz quantum cascade laser spectroscopy," Phys. Rev. X **4**, 021006 (2014).

7. S. Barbieri, J. Alton, H. E. Beere, J. Fowler, E. H. Linfield, and D. A. Ritchie, "2.9 THz quantum cascade lasers operating up to 70 K in continuous wave," Appl. Phys. Lett. **85**, 1674–1676 (2004).
8. P. Gellie, S. Barbieri, J.-F. Lampin, P. Filloux, C. Manquest, C. Sirtori, I. Sagnes, S. P. Khanna, E. H. Linfield, A. G. Davies, H. Beere, and D. Ritchie, "Injection-locking of terahertz quantum cascade lasers up to 35GHz using RF amplitude modulation," Opt. Express **18**, 20799–20816 (2010).
9. A. Bartels, D. Heinecke, and S. A. Diddams, "Passively mode-locked 10 GHz femtosecond Ti:sapphire laser," Opt. Lett. **33**, 1905–1907 (2008).
10. O. Kliebisch, D. C. Heinecke, and T. Dekorsy, "Ultrafast time-domain spectroscopy system using 10 GHz asynchronous optical sampling with 100 kHz scan rate," Opt. Express **24**, 29930–29940 (2016).
11. G. Klatt, R. Gebs, C. Janke, T. Dekorsy, and A. Bartels, "Rapid-scanning terahertz precision spectrometer with more than 6 THz spectral coverage," Opt. Express **17**, 22847–22854 (2009).
12. L.-S. Ma, M. Zucco, S. Picard, L. Robertsson, and R. S. Windeler, "A new method to determine the absolute mode number of a mode-locked femtosecond-laser comb used for absolute optical frequency measurements," IEEE J. Sel. Top. Quantum. Electron. **9**, 1066–1071 (2003).
13. S. Barbieri, P. Gellie, G. Santarelli, L. Ding, W. Maineult, C. Sirtori, R. Colombelli, H. Beere, and D. Ritchie, "Phase-locking of a 2.7-THz quantum cascade laser to a mode-locked erbium-doped fibre laser," Nat. Photonics **4**, 636–640 (2010).
14. W. J. Riley, *Handbook of Frequency Stability Analysis* (National Institute of Standards and Technology, 2008).
15. H. Pickett, R. Poynter, E. Cohen, M. Delitsky, J. Pearson, and H. Müller, "Submillimeter, millimeter, and microwave spectral line catalog," J. Quant. Spectrosc. Radiat. Transfer **60**, 883–890 (1998).
16. G. Moruzzi, M. Winnewisser, I. Mukhopadhyay, and F. Strumia, *Microwave, Infrared, and Laser Transitions of Methanol Atlas of Assigned Lines from 0 to 1258 cm−1* (Taylor & Francis, 1995).
17. T. Yasui, K. Hayashi, R. Ichikawa, H. Cahyadi, Y.-D. Hsieh, Y. Mizutani, H. Yamamoto, T. Iwata, H. Inaba, and K. Minoshima, "Real-time absolute frequency measurement of continuous-wave terahertz radiation based on dual terahertz combs of photocarriers with different frequency spacings," Opt. Express **23**, 11367–11377 (2015).