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Frequency stabilization of a terahertz quantum-cascade laser to the Lamb dip of a molecular absorption line

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Abstract: We demonstrate the frequency stabilization of a terahertz quantum-cascade laser (QCL) to the Lamb dip of the absorption line of a D₂O rotational transition at 3.3809309 THz. To assess the quality of the frequency stabilization, a Schottky diode harmonic mixer is used to generate a downconverted QCL signal by mixing the laser emission with a multiplied microwave reference signal. This downconverted signal is directly measured by a spectrum analyzer showing a full width at half maximum of 350 kHz, which is eventually limited by high-frequency noise beyond the bandwidth of the stabilization loop.

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1. Introduction

Terahertz (THz) spectroscopy is used in several research fields such as astronomy, atmospheric research and metrology as well as in laboratory spectroscopy [1–3]. A large number of atomic fine-structure lines and molecular rotational lines is located in the THz range. Spectroscopic methods like heterodyne spectroscopy require sources with small linewidths in the MHz or sub-MHz range with a sufficient power level. In heterodyne spectroscopy, quantum-cascade lasers (QCLs) are used as local oscillators (LO) as they offer output power in the range of several milliwatts above 3 THz, where other available sources offer output powers in the microwatt range [4–8]. The theoretical linewidth limit of QCLs, due to spontaneous emission (Schawlow-Townes), lies in the sub-kHz range [9]. However, their practically achievable linewidth is governed by the technical limitations of the respective setup. Due to instabilities of the operating current and temperature as well as other noise sources such as vibrations and optical feedback, the practical linewidth of QCLs is much larger and can exceed several ten MHz [10]. Several methods to control and stabilize the emission frequency have been realized, e.g. phase locking to a multiplied microwave source or frequency comb [11,12] or locking to a THz gas laser [13]. Another well established method is the stabilization to a molecular or atomic absorption line based on wavelength modulation spectroscopy. This has also been demonstrated for QCLs, where the frequency stability is indirectly measured by converting the proportional-integral-derivative (PID) error signal into a frequency scale [14,15]. The stabilization to the Lamb dip of a molecular

transition has been shown for a mid-infrared QCL in Refs. [16,17]. The general feasibility of saturation spectroscopy with THz QCLs has been demonstrated in [18,19]. In this work, we demonstrate the frequency stabilization of a THz QCL to the Lamb dip of a D₂O rotational transition by means of a wavelength modulation technique. Due to the much smaller linewidth of the Lamb dip compared to the unsaturated transition, an improvement in frequency stability by an order of magnitude can be expected. By downconverting the THz signal with the help of a Schottky diode mixer and a multiplied microwave reference, we are able to directly measure the width of the signal with a spectrum analyzer. This allows for an assessment of the performance of the Lamb dip stabilization approach.

2. Experimental setup

The experimental configuration is shown in Fig. 1. The QCL used in our work is based on a 3-THz active-region structure [20] with a single-plasmon waveguide [21]. It is mounted inside a He flow cryostat (Oxford, Optistat CF-V), and its temperature is stabilized to 40 K. The QCL beam exits the cryostat through a high-density polyethylene (HDPE) window and is focused by a polymethylpentene (TPX) lens with a focal length of 45 mm into a 35-cm-long D₂O gas cell. To minimize the optical feedback and to redirect the probe signal to the photoconductive Ge:Ga detector, a $\lambda/4$ plate and a wire grid polarizer are used. Additionally, an aperture after the $\lambda/4$ plate is used to suppress higher-order spatial modes and to minimize optical feedback. The shorter gas cell as well as better filtering of spatial modes led to significantly reduced interferences caused by optical feedback compared to our previous work [15,22]. A shorter gas cell also allows for a better focus and higher power density. The Lamb dip of the 3.3809-THz absorption line of D₂O [23] was used for this experiment as shown in Fig. 2. This transition is one of the strongest for D₂O in the 3 to 4 THz range and close to the 3.5 THz transition of the OH radical, which is of importance in atmospheric chemistry [24]. A lock-in amplifier with an integrated PID controller (Zurich Instruments, UHF) is used to obtain the 1f signal at 1-MHz modulation frequency, which corresponds to the derivative of the direct signal. This is used by the PID controller to stabilize the QCL frequency to the zero crossing of the 1f signal. The locking range of the stabilization loop is defined by the linear range of the 1f signal indicated by the grey area in Fig. 2, which corresponds to a range of 40 μ A equivalent to 750 kHz. To establish the lock, the QCL frequency needs to stay within these 750 kHz for the capture time of the control loop. Additionally to the Millikelvin temperature control (40 K \pm 2 mK), a star-shaped electrical grounding was used to reduce ground loops, and an optimized detector readout circuitry was employed for increased bandwidth and reduced pick-ups.

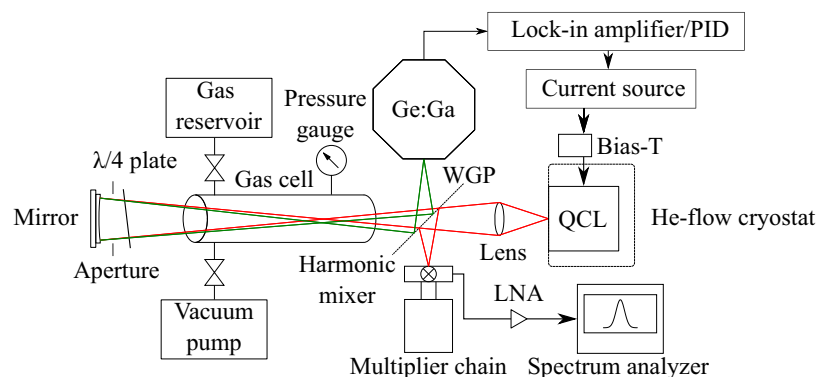


Fig. 1. Experimental setup for frequency stabilization. WGP: wire grid polarizer, LNA: low-noise amplifier, QCL: quantum-cascade laser.

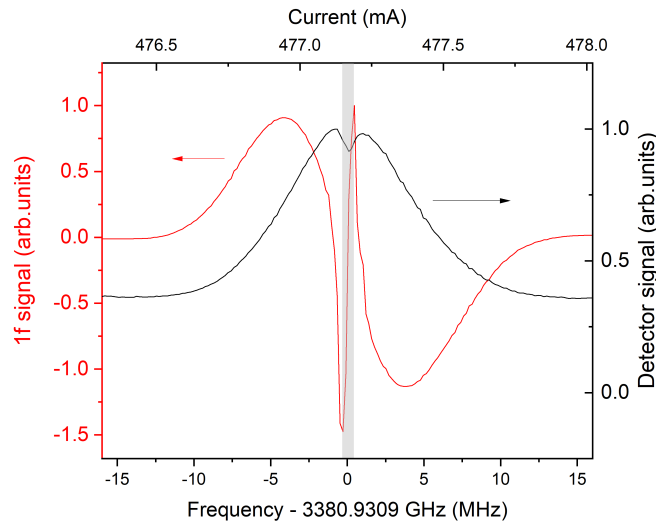


Fig. 2. 1f (red) and direct-detection (black) absorption spectrum of the rotational transition of D_2O at 3380.9309 GHz at 0.75 Pa pressure. The line corresponds to the $D_2O(4,4,0) \leftarrow D_2O(3,3,1)$ transition [23]. The Lamb dip is clearly visible in both signals. The grey area indicates the locking range of the PID loop of 750 kHz.

A small portion of the pump signal is redirected onto the harmonic Schottky diode mixer [25]. We downconvert the THz QCL signal to a range of 100 to 200 MHz by mixing it with the 6th harmonic of a 563-GHz multiplier chain. The latter is generated by an amplifier multiplier chain from a microwave source at 11.74 GHz (VDI, Tx 249), referenced to an internal 10 MHz oscillator (Wenzel Associates 501-04609A). This downconverted intermediate-frequency (IF) signal is further amplified and measured by a spectrum analyzer to obtain the spectral width of the QCL emission.

3. Results and discussion

Figure 2 displays the D_2O absorption line as measured by sweeping the QCL current. The black line shows the Ge:Ga detector signal as directly recorded with a data acquisition module (National Instruments, NI-9239), while the red line shows the 1f signal after demodulation by the lock-in amplifier. The frequency tuning coefficient of the QCL is 18.71 MHz/mA and the total measurement time in Fig. 2 is 25 s.

Once the lock is engaged, the frequency stability of the QCL increases significantly. Figure 3(a) depicts the time-dependent frequency deviation of the IF signal of the laser modulated at 1 MHz with a modulation amplitude of 65 μA , which corresponds to a tuning of 1.2 MHz, over a time interval of 25 s. Each spectrum was integrated over 100 ms. In the first 12 s (A), the laser is stabilized without any drift. After about 12 s (B), the laser setpoint is slowly drifting outside the locking range of the PID controller, and an increase in the frequency fluctuations appears. In the third part (C), the active stabilization is turned off, and the frequency fluctuations of the unstabilized QCL are observed. The increase of frequency stability can also be seen in Fig. 3(b), where the maximum of the spectral power of the IF signal of the unstabilized and stabilized QCL emission with respect to the stable multiplier reference is recorded over 10 s. The stabilized signal shows the expected narrow central peak along with sidepeaks caused by the 1-MHz modulation, while the unstabilized signal is broadened due to frequency fluctuations.

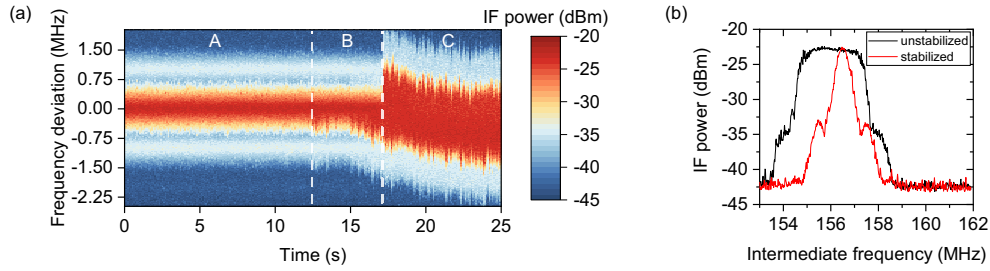


Fig. 3. (a) Time-dependent frequency deviation of the IF signal. The integration time for each slice is 100 ms. A: the QCL is stabilized without any drift, B: the setpoint of the stabilized laser is drifting outside the locking range, and C: the stabilization is turned off and the frequency fluctuations of the free-running laser are visible. (b) Maximum-hold measurement of the IF power for the unstabilized and stabilized case recorded over 10 s.

The power spectrum of the stabilized QCL at different modulation amplitudes measured by the spectrum analyzer is presented in Fig. 4(a). These measurements were performed to quantify the influence of the modulation amplitude on the stabilized laser linewidth. The signal was integrated for 78 s to obtain a better signal-to-noise ratio in the side lobes. The modulation depths range from 0.6 to 1.8 MHz. The central peaks have a full width at half maximum (FWHM) of about 350 kHz for all modulation depths. The sidepeaks are caused by the 1-MHz modulation and are located at the center frequency of the main peak $\pm N \times 1$ MHz. To identify the individual peak areas, each peak has been fitted by a Gaussian. Note that although overlapping on a logarithmic scale the peaks would appear well separated on a linear power scale. The area of the central peak decreases linearly with the modulation depth, while the areas of the sidepeaks increase linearly as more power is shifted from the central peak to the sidepeaks (Fig. 4(b)). Although the power ratio changes slightly, the widths of the central peak and the sidepeaks are unaffected by the modulation depths in the exploited range.

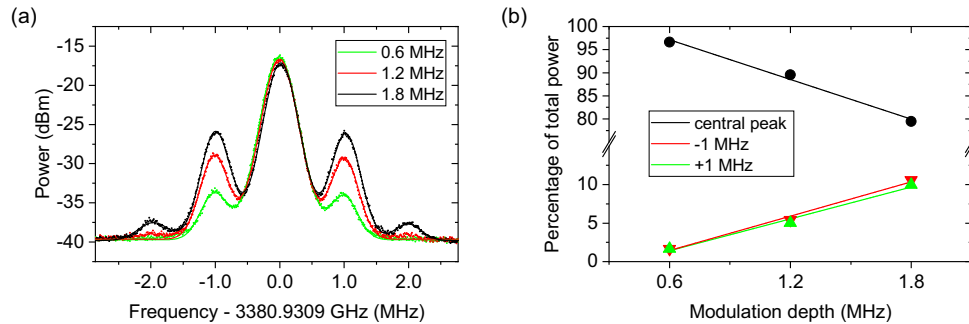


Fig. 4. (a) Power spectrum of the stabilized and downconverted QCL signal at different modulation depths. The central peak has a FWHM of 350 kHz for all modulation depths, integrated over 78 s. Sidepeaks emerge at multiples of the modulation frequency of 1 MHz. The solid lines represent a multi-peak fit to precisely calculate the peak areas. (b) Peak area of the central peak and first-order peaks of (a). Solid lines represent linear fits. The peak area of the central peak decreases linearly with the modulation depth, while the areas of the sidepeaks increase.

Aside from the slope of the 1f signal of the Lamb dip, the performance of the stabilization is limited by the loop-bandwidth and the sensitivity of the detector. The loop bandwidth is determined by the bandwidth of the detector (1.3 MHz), and the required lock-in integration time.

Typical values as used for Fig. 4 are 9.3 and 1.8 kHz for the lock-in bandwidth, i.e. for the 1f signal, and the PID loop, respectively.

Fig. 5(a) shows the spectral power density of the frequency noise of the QCL (1f signal) and the β -separation line [26], which is used to calculate the width of the emission, resulting in an FWHM of 16 kHz. The width of the QCL emission can also be derived from the frequency fluctuations during the stabilization by scaling the linear range of the 1f signal to its frequency range as shown in Fig. 5(b). This procedure yields an FWHM of the frequency distribution of 28 kHz. Although both in-loop methods yield similar results, the linewidth differs significantly from the 350-kHz width measured directly with the spectrum analyzer. This significantly larger linewidth was unexpected and is most likely explained by the insufficient loop bandwidth to detect and counteract high-frequency electronic noise, which appears to be intrinsic to the QCL structure. An explanation for high-frequency noise might be unavoidable growth imperfections and defects, which are known to cause flicker noise [27]. To eliminate errors due to one specific instrument, these measurements have been performed with several different lock-in amplifiers (Zurich Instruments UHF, Toptica Digilock 110) and QCL current drivers (QCL 1000 LAB, QubeCL). There have been no notable changes in performance between the different instruments. Therefore, we conclude that a substantially larger detector bandwidth and sensitivity would be necessary to improve the stabilization with respect to high-frequency electronic noise.

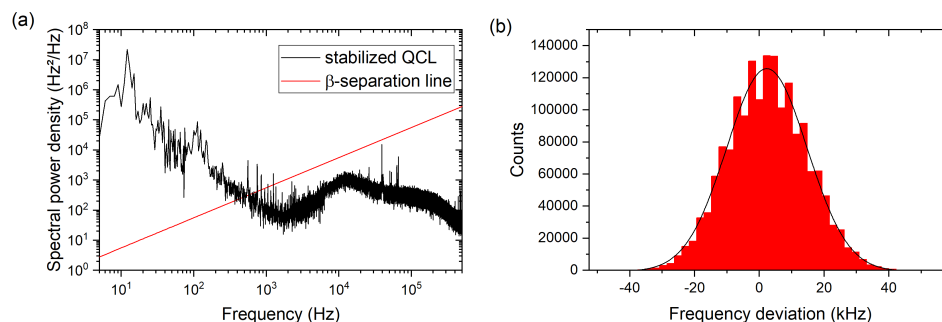


Fig. 5. (a) Spectral power density of the frequency noise of the stabilized QCL signal and the β -separation line. Calculating the width of the laser emission with the β -separation method yields an FWHM of 16 kHz. (b) Histogram of the stabilized QCL frequency deviation, indicating an FWHM of about 28 kHz.

4. Conclusion

A QCL was locked to the Lamb dip of a D₂O rotational at 3.38 THz transition by using a PID controller to adjust the QCL current based on the 1f signal generated by a lock-in amplifier. The width of the stabilized laser measured with respect to a multiplier reference is 350 kHz. The results indicate that the linewidth of the QCL is not limited by spontaneous emission, but by the intrinsic electronic noise of the QCL. We expect to achieve the required larger loop bandwidth by using a detector with a higher specific detectivity such as a hot-electron bolometer.

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Disclosures. The authors declare no conflicts of interest.

Data availability. The data presented in this paper is available from the authors upon reasonable request.

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