# Submegahertz frequency stabilization of a terahertz quantum cascade laser to a molecular absorption line

H. Richter,[1,a)] S. G. Pavlov,[1] A. D. Semenov,[1] L. Mahler,[2] A. Tredicucci,[2] H. E. Beere,[3] D. A. Ritchie,[3] and H.-W. Hübers[1,4]

[1]*German Aerospace Center (DLR), Institute of Planetary Research, Rutherfordstr. 2, 12489 Berlin, Germany*
[2]*NEST CNR-INFM and Scuola Normale Superiore, Piazza dei Cavalieri 7, 56126 Pisa, Italy*
[3]*Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 0HE, United Kingdom*
[4]*Institut für Optik und Atomare Physik, Technische Universität Berlin, Hardenbergstraße 36, 10623 Berlin, Germany*

(Received 3 November 2009; accepted 27 January 2010; published online 19 February 2010)

The frequency of a terahertz quantum-cascade laser is stabilized to the absorption line of methanol gas at a frequency of 2.55 THz. The method is based on frequency modulation of the laser emission across the absorption line. The resulting derivativelike signal is used as an error signal for a control loop that keeps the laser frequency at maximum absorption. The unstabilized laser that is operated in a pulse tube cooler has frequency fluctuations of 15 MHz, which are reduced to 300 kHz with the control loop in action. The line shape of the locked signal is Gaussian. © *2010 American Institute of Physics*. [doi:10.1063/1.3324703]

Since the demonstration of a terahertz (THz) quantum cascade laser (QCL) in 2002 (Ref. 1) its performance was rapidly improved with respect to output power, operation temperature, and frequency coverage.[2] The principal applicability of these lasers for THz imaging[3] and high resolution molecular spectroscopy[4] has been demonstrated. Other emerging applications are in security and biomedicine or as local oscillator (LO) in a THz heterodyne spectrometer. Despite important steps toward a QCL-based LO in the past few years,[5–8] heterodyne spectroscopy has not been demonstrated with a QCL as LO, mainly because its free-running linewidth is too large. For high resolution spectroscopy sub-MHz accuracy and linewidth of the LO are required. Environmental effects such as bias current and temperature variations affect the QCL frequency. Previous measurements have shown, that a free running THz-QCL has a full width at half maximum (FWHM) linewidth of about 20 KHz measured within about 4 ms.[7,9] But for longer integration times the linewidth exceeds 10 MHz.[9] In order to control the QCL frequency and linewidth to sub-MHz accuracy it has to be locked to some external reference. Locking to the frequency of a THz gas laser[10,11] as well as to that of a multiplied microwave source[12,13] has been demonstrated. The FWHM was several kilohertz and less than 100 Hz, respectively, and the lock condition could be maintained indefinitely. However, phase locking to a reference line from a THz gas laser is neither a very practical solution nor a very versatile technique since gas lasers are relatively bulky and appropriate laser lines become sparse above 3 THz. Frequency locking to the emission from a multiplied microwave source becomes increasingly difficult with increasing frequency due to the decrease of power from these sources.

An alternative stabilization technique is based on an atomic or molecular resonance serving as frequency reference. This technique was developed shortly after the invention of the laser.[14,15] Subsequently it was applied to a variety of laser types and it is now a well-established technique in most parts of the electromagnetic spectrum. For example, for mid-infrared QCLs a dithering-free locking technique to the side of a rovibrational resonance of $N_2O$ is described in Ref. 16 and lamb-dip frequency stabilization is described in Ref. 17.

Here we report on the frequency stabilization of a THz QCL relative to the center frequency of a methanol gas absorption line. The difference between the frequency of the absorption maximum and the QCL frequency was used to generate an error signal for the stabilization. Sub-MHz linewidth and accuracy have been achieved. This approach overcomes the shortcomings of the other frequency locking schemes, because it requires only an additional detector and a small gas absorption cell while being applicable even at the highest THz frequencies due to the rich absorption spectra of molecules such as $CH_3OH$ or $H_2O$.

A schematic representation of the experiment is shown in Fig. 1. The QCL is based on a GaAs/AlGaAs superlattice and a plasmon-type waveguide operating at 2.5 THz with a Fabry–Pérot cavity that is 240 $\mu$m wide and 2.5 mm long.[18]

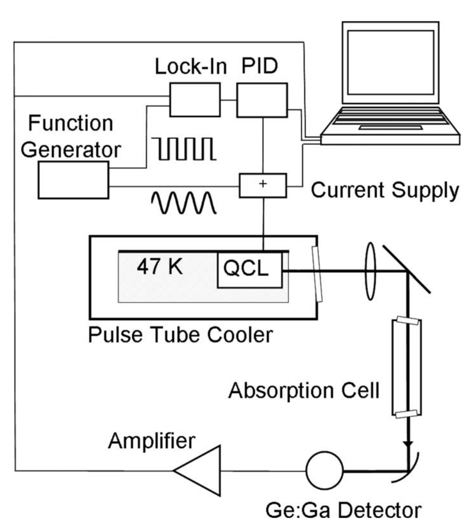


FIG. 1. Scheme of the experimental setup for the frequency stabilization.

[a)]Electronic mail: heiko.richter@dlr.de.

 

The laser is soldered to a copper plate, wire bonded, and mounted onto the first cold stage of a Gifford–McMahon-type pulse tube cooler (PTC) (model PTD-406 from TransMIT GmbH, Gießen, Germany). During operation of the QCL the temperature of the cold stage rises from 39 to 47 K due to the increased heat load of about 7 W from the QCL. In continuous wave mode the laser provides a maximum output power of about 2 mW and the threshold current is 585 mA at 46 K.

The diverging QCL beam was focused with a TPX lens resulting in a Gaussian shaped beam with an $M^2$ value of about 1.2.[8] The radiation was guided through a 52 cm long absorption cell. The vacuum windows of the cell were tilted in order to avoid standing waves and the pressure in the cell was measured with a capacitive manometer. The cell was filled with $^{12}CH_3OH$ at a pressure of 1–2 hPa. Methanol was chosen, because it has many absorption lines in the frequency range covered by the QCL. The particular absorption line that was used as a reference for the stabilization is a transition at 2.55025 THz. The signal transmitted through the absorption cell was detected with a liquid helium cooled Ge:Ga photoconductor, followed by a low noise amplifier. The output signal of this amplifier was either directly read out or it was further amplified by a lock-in amplifier.

The bias current for the QCL was supplied from a battery connected to a home-made current supply, which allows the bias current to be controlled by three independent input signals. One of these inputs was used for setting the dc operation point, the second for applying a sinusoidal modulation signal, and the third for adding the control signal of the proportional-integral-derivative (PID) control loop. A computer interfaced with a field programmable gate array (FPGA) was used for data acquisition. The lock-in amplifier and the PID control loop were implemented in the FPGA. A dc bias current was applied for setting the center frequency. The current of the QCL was modulated by adding a small sinusoidal current from a function generator. The frequency and the magnitude of the modulation current could be varied from 0 to 10 kHz and from 0 to 1 mA, respectively. The shape of the absorption signal was registered with the lock-in amplifier and the current modulation frequency as reference. By this means the tilted baseline that occurs due to changing laser power if the direct absorption signal is recorded can be removed as well as $1/f$ noise. The signal is approximately the first derivative of the shape of the absorption line with the baseline crossing at the minimum of the absorption line (Fig. 2). The PID control loop is based on this derivativelike signal, which is used to generate the error signal of the loop. Locking is accomplished by implementing a feedback, which keeps the QCL frequency at the center of the absorption line where the derivativelike signal crosses the baseline. The dc offset caused by the slope of the transmission signal is compensated in order to yield a zero error signal. The complete controlling and processing was implemented into a control software allowing the adjustment of all settings and monitoring the error signal during operation.

For evaluation the voltage variations of the error signal need to be translated into frequency variations. First the voltage variations are transformed into current variations using the current-voltage relationship of the error signal in the locking range around the center of the absorption line, where the error signal is almost linear. The dependence of the laser frequency on the current was determined by measuring the absorption spectrum of methanol as a function of the laser current between 550 and 980 mA and at fifty temperature values between 39 and 48 K. The resulting spectra have up to six absorption lines whose positions are a function of current and temperature of the QCL. The pattern of the absorption lines is fingerprint-like and allows the identification of the lines whose precise frequencies are known from a reference spectrum measured with a Fourier transform spectrometer or from the literature.[19] The positions of the absorption lines are fitted assuming a linear dependence of the QCL frequency on current and temperature in the range of interest. This is justified because a similar dependence has been found by difference frequency measurements of other laser modes from the same QCL with respect to the emission of a THz gas laser.[20] The absorption lines were fitted with a precision of better than $3\times10^{-6}$. The fitting procedure yields a frequency tuning of $+(3.5\pm0.4)$ MHz/mA and temperature tuning of $-(82\pm18)$ MHz/K. These values are in good agreement with those measured at 20 K (+4 to +8 MHz/mA and −100

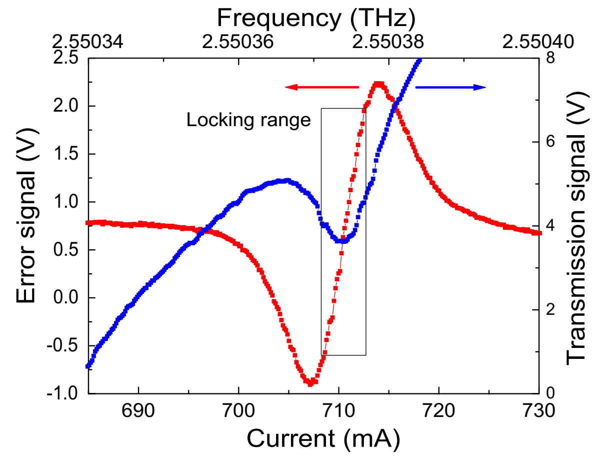


FIG. 2. (Color online) Absorption line of methanol at 2.55 THz and 1.7 hPa. The transmission signal is measured in direct detection and the error signal measured with the lock-in amplifier and the frequency of the QCL current modulation as reference. The QCL is locked to the center of the derivative-like error signal. The locking range is indicated by the rectangular box.

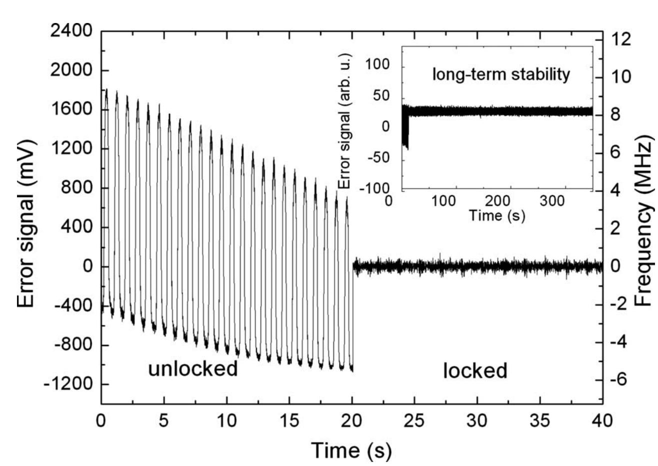


FIG. 3. Error signal in the unlocked and in the locked state of the PID control loop. The control loop was activated after 20 s. The variations in the unlocked state are caused by temperature and current fluctuations in the QCL. The large variation resembles the temperature cycle of the pulse tube cooler. The inset shows the long term stability with the control loop activated after 12 s.

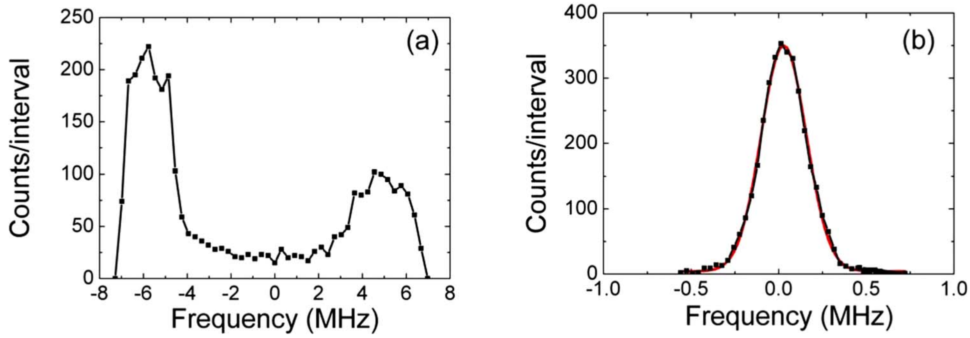


FIG. 4. (Color online) Number of counts in a given frequency interval when the QCL is operating in the locked (a) and unlocked (b) state for 10 s. For longer time intervals the peaks in the left diagram become much broader, while the width and shape of the peak in the right diagram remains unchanged. The solid line in (b) is a Gaussian fit to the measurement.

MHz/K).[20] With the voltage-current-frequency dependence determined by this way the voltage is converted into frequency.

The result of the stabilization is shown in Fig. 3. In the unlocked state, the derivativelike signal shows peak-to-peak variations of 15 MHz at a frequency of 1.2 Hz as well as a slow drift component. The 1.2 Hz variation is related to the temperature variation on the first cold plate of the PTC, which is 0.1 K as measured with a Pt-100 thermistor. The thermal drift is due to current heating of the QCL. By monitoring the position of an absorption line as a function of time the associated frequency drift was readily observable. In the locked state the 1.2 Hz component and the thermal drift are completely eliminated. The peak-to-peak frequency variation in the locked state is approximately 600 kHz. In order to determine linewidth and lineshape the frequency ordinate is subdivided into 300 kHz wide intervals (unlocked state) and 34 kHz wide intervals (locked state) and the number of frequency values in each interval is counted. This number is plotted as a function of frequency in Fig. 4. In the unlocked state two peaks with a distance of 10 MHz appear. They are caused by the 1.2 Hz disturbance of the PTC. With the temperature gradient of $-82$ MHz/K this corresponds to a temperature difference of 0.12 K, which is in agreement with the 0.1 K variations measured on the first cold plate at a slightly different position from the QCL. The FWHM of each of the two peaks is approximately 3 MHz. This is caused by current and temperature induced instabilities. In the locked state, only one peak with a FWHM of $(300 \pm 35)$ kHz exists. The uncertainty is determined by the uncertainty in the determination of the current tuning. The achieved linewidth is sufficient for many applications. For example the frequency resolution of a heterodyne receiver with an acousto-optical backend spectrometer is 1.5 MHz.[21] With a 300 kHz linewidth of the LO this increases marginally to 1.53 MHz.

In summary, the emission of a QCL operating at 2.55 THz has been locked to an absorption line of methanol gas. The unstabilized QCL has thermal drift induced and short-term frequency fluctuations of 15 MHz. These are reduced to 300 kHz in a Gaussian shaped line profile with a PID control loop. The stabilization scheme is robust and versatile, because it requires only an additional detector and a small gas absorption cell while being applicable even at the highest THz frequencies due to the rich absorption spectra of molecules such as $CH_3OH$ or $H_2O$.

The authors would like to thank M. Greiner-Bär for technical assistance.

[1]R. Köhler, A. Tredicucci, F. Beltram, H. E. Beere, E. H. Linfield, A. G. Davies, D. A. Ritchie, R. C. Iotti, and F. Rossi, Nature (London) **417**, 156 (2002).
[2]B. S. Williams, Nat. Photonics **1**, 517 (2007).
[3]A. W. M. Lee, Q. Qin, S. Kumar, B. S. Williams, Q. Hu, and J. L. Reno, Appl. Phys. Lett. **89**, 141125 (2006).
[4]H.-W. Hübers, S. G. Pavlov, H. Richter, A. D. Semenov, L. Mahler, A. Tredicucci, H. E. Beere, and D. A. Ritchie, Appl. Phys. Lett. **89**, 061115 (2006).
[5]H.-W. Hübers, IEEE J. Sel. Top. Quantum Electron. **14**, 378 (2008).
[6]J. R. Gao, J. N. Hovenier, Z. Q. Yang, J. J. A. Baselmans, A. Baryshew, M. Hajenius, T. M. Klapwijk, A. J. L. Adam, T. O. Klaassen, B. S. Williams, S. Kumar, Q. Hu, and J. L. Reno, Appl. Phys. Lett. **86**, 244104 (2005).
[7]H.-W. Hübers, S. G. Pavlov, A. D. Semenov, R. Köhler, L. Mahler, A. Tredicucci, H. E. Beere, D. A. Ritchie, and E. H. Linfield, Opt. Express **13**, 5890 (2005).
[8]H. Richter, A. D. Semenov, S. Pavlov, L. Mahler, A. Tredicucci, K. Il'in, M. Siegel, and H.-W. Hübers, Appl. Phys. Lett. **93**, 141108 (2008).
[9]A. Barkan, F. K. Tittel, D. M. Mittleman, R. Dengler, P. H. Siegel, G. Scalari, L. Ajili, J. Faist, H. E. Beere, E. H. Linfield, A. G. Davies, and D. A. Ritchie, Opt. Lett. **29**, 575 (2004).
[10]A. L. Betz, R. T. Boreiko, B. S. Williams, S. Kumar, Q. Hu, and J. L. Reno, Opt. Lett. **30**, 1837 (2005).
[11]A. A. Danylov, T. M. Goyette, J. Waldman, M. J. Coulombe, A. J. Gatesman, R. H. Giles, W. D. Goodhue, X. Qian, and W. E. Nixon, Opt. Express **17**, 7525 (2009).
[12]D. Rabanus, U. U. Graf, M. Philipp, O. Ricken, J. Stutzki, B. Vowinkel, M. C. Wiedner, C. Walther, M. Fischer, and J. Faist, Opt. Express **17**, 1159 (2009).
[13]P. Khosropanah, A. Baryshev, W. Zhang, W. Jellema, J. N. Hovenier, J. R. Gao, T. M. Klapwijk, D. G. Paveliev, B. S. Williams, Q. Hu, J. L. Reno, B. Klein, and J. L. Hesler, Opt. Lett. **34**, 2958 (2009).
[14]A. D. White, E. I. Gordon, and E. F. Labuda, Appl. Phys. Lett. **5**, 97 (1964).
[15]P. H. Lee and M. L. Skolnick, Appl. Phys. Lett. **10**, 303 (1967).
[16]R. W. Williams, J. F. Kelly, J. S. Hartman, S. W. Sharpe, M. S. Taubman, J. L. Hall, F. Capasso, C. Gmachl, D. L. Sivco, J. N. Baillargeon, and A. Y. Cho, Opt. Lett. **24**, 1844 (1999).
[17]S. Borri, S. Bartalini, I. Galli, P. Cancio, G. Giusfredi, D. Mazzotti, A. Castrillo, L. Gianfrani, and P. De Natale, Opt. Express **16**, 11637 (2008).
[18]L. Mahler, A. Tredicucci, R. Köhler, F. Beltram, H. E. Beere, E. H. Linfield, and D. A. Ritchie, Appl. Phys. Lett. **87**, 181101 (2005).
[19]G. Moruzzi, F. Strumia, P. Carnesecchi, B. Carli, and M. Carlotti, Infrared Phys. **29**, 47 (1989).
[20]H.-W. Hübers, S. G. Pavlov, A. D. Semenov, L. Mahler, A. Tredicucci, H. E. Beere, and D. A. Ritchie, Proc. SPIE **6275**, 62750H (2006).
[21]J. Horn, O. Siebertz, F. Schmülling, C. Kunz, R. Schieder, and G. Winnewisser, Exp. Astron. **9**, 17 (1999).