Frequency locking of single-mode 3.5-THz quantum cascade lasers using a gas cell

Y. Ren,[1,2,3,a)] J. N. Hovenier,[1] M. Cui,[4] D. J. Hayton,[4] J. R. Gao,[1,4,a)] T. M. Klapwijk,[1] S. C. Shi,[2] T.-Y. Kao,[5] Q. Hu,[5] and J. L. Reno[6]

[1]Kavli Institute of NanoScience, Delft University of Technology, Lorentzweg 1, 2628 CJ Delft, The Netherlands
[2]Purple Mountain Observatory (PMO), Chinese Academy of Sciences, 2 West Beijing Road, Nanjing, JiangSu 210008, China
[3]Graduate School, Chinese Academy of Sciences, 19A Yu Quan Road, Beijing 100049, China
[4]SRON Netherlands Institute for Space Research, Sorbonnelaan 2, 3584 CA Utrecht, The Netherlands
[5]Department of Electrical Engineering and Computer Science, Massachusetts Institute of Technology (MIT), Cambridge, Massachusetts 02139, USA
[6]Center for Integrated Nanotechnologies, Sandia National Laboratories, Albuquerque, New Mexico 87185-0601, USA

We report frequency locking of two 3.5-THz third-order distributed feedback (DFB) quantum cascade lasers (QCLs) by using methanol molecular absorption lines, a proportional-integral-derivative controller, and a NbN bolometer. We show that the free-running linewidths of the QCLs are dependent on the electrical and temperature tuning coefficients. For both lasers, the frequency locking induces a similar linewidth reduction factor, whereby the narrowest locked linewidth is below 18 kHz with a Gaussian-like shape. The linewidth reduction factor and the ultimate linewidth correspond to the measured frequency noise power spectral density. © 2012 American Institute of Physics. [doi:10.1063/1.3679620]

A phase- or frequency-stabilized solid-state source is of crucial importance for its application as local oscillator (LO) for high-resolution heterodyne spectroscopy in the terahertz (THz) frequency range for astronomical and atmospheric research, particularly from space. As solid-state sources above 2 THz, quantum cascade lasers (QCLs) have shown great advantages, based on their broad frequency coverage, single-mode emission, and high output power.[1] It has been shown theoretically[2,3] and experimentally[4,5] that THz QCLs can have narrow intrinsic linewidths of below tens of kHz. However, caused by the fluctuations in the electrical bias and in the operating temperature, the practical linewidth of a free-running THz QCL measured over a long period (>1 s) is usually much broader, typically greater than 1 MHz.[4,6]

Phase locking of a THz QCL to an external up-converted reference source has been demonstrated,[6,7] where both the phase and frequency of the laser radiation are stabilized. However, in this phase locking approach, an additional THz reference source is required, that can be difficult to implement at frequencies above 3 THz due to the lack of suitable sources. Alternatively, phase locking of a 2.5 THz QCL to a frequency comb using a GaAs photomixer was developed recently.[8] The advantage of this approach is the possibility of using a room temperature detector. Additionally, the technique can be applied, in principle, to any frequency. In some applications such as a heterodyne interferometer,[9] phase locking of multiple LOs to a common reference is essential. However, for an observation based on a single telescope, frequency locking, where the LO's average frequency is stabilized, but where its linewidth remains intrinsic, is often sufficient. A frequency stabilization scheme based on a molecular line as reference frequency was demonstrated for a 2.5 THz QCL.[10] In this case, the QCL's frequency was stabilized, resulting in a full width at half maximum (FWHM) linewidth of 300 kHz. It is worth noting that the locked linewidth is at least an order of magnitude greater than the intrinsic linewidth,[2–5] suggesting that the full potential of this technique was not achieved yet. However, this approach is simple and robust because it requires in essence only a gas cell unit and a direct power detector which does not have to be operated at very low temperature.

Here, we perform the frequency stabilization measurements using a methanol (CH_3OH) absorption line, but with two advanced, frequency controllable distributed feedback (DFB) QCLs (Ref. 11) at much higher frequencies. Furthermore, we apply a superconducting NbN bolometer as a power detector to monitor the radiation signal after a gas cell. We report not only a much narrower locked linewidth, but also address the device dependence on the free running linewidth and line shape.

The QCLs used are two third-order DFB QCLs. The QCL lasing at 3.45 THz is labeled as "laser A" and the other at 3.35 THz as "laser B." The two lasers, based on a 10-μm thick active region, consist of 27 periods of gratings, but with slightly different periodicity. The lasers are designed and fabricated by the MIT group. As demonstrated previously,[12] by using the third-order periodic structure with strong refractive index contrast gratings, not only can single mode emission be achieved, but also a less divergent, single spot far-field beam. Each QCL is mounted on the second stage of a pulse tube cryocooler ($\sim$12 K under load) without further temperature stabilization.

The measurement setup is schematically described in Fig. 1. The QCL beam is first focused with a high-density

a)Authors to whom correspondence should be addressed. Electronic addresses: y.ren@tudelft.nl and j.r.gao@tudelft.nl.
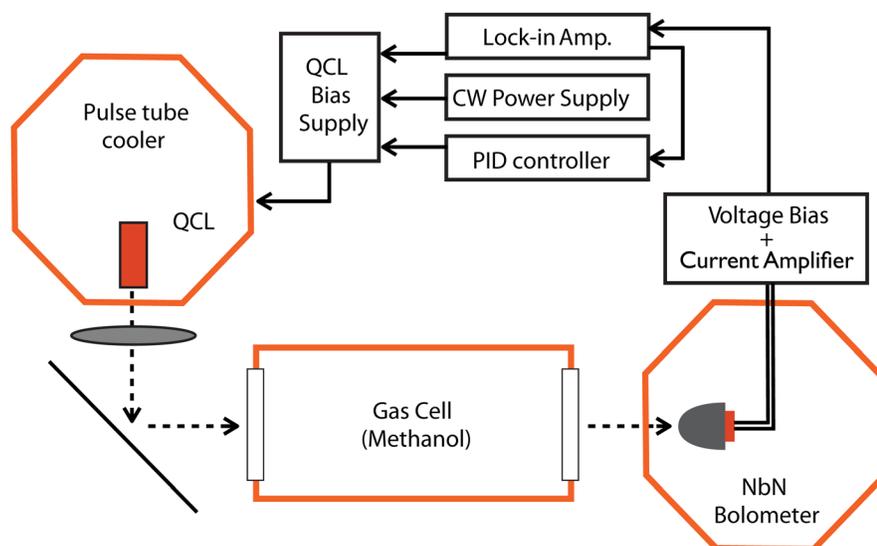
FIG. 1. (Color online) Schematic of the frequency locking measurement setup.

polyethylene (HDPE) lens (f = 26.5 mm), and then guided through a gas cell by a 13-μm thick Mylar beam splitter. The gas cell, containing methanol gas at room temperature, is a 41-cm long cylinder with two 2-mm thick HDPE windows. Methanol gas is chosen since it contains abundant absorption lines around the QCL frequencies. The transmitted signal through the gas cell is monitored by a superconducting NbN bolometer operated at liquid helium temperature as a direct detector, whereby we benefit from a NEP of $10^{-12} - 10^{-13}$ W/Hz$^{1/2}$ (Ref. [13]) and a fast response ($\sim$40 ps).

To perform frequency locking, we apply a summing bias circuit that allows combining three input signals to independently control the bias voltage of a laser.[10] The first input is a standard DC bias voltage, which sets the operating point of the laser. The second input is an AC sinusoidal modulation signal at around 1 kHz with relatively small amplitude ($<$0.01% of the DC bias). By feeding the detector output current to a lock-in amplifier with the AC modulation signal as reference frequency, the derivative signal of the absorption profile is obtained. The third one is the control signal from a feedback loop. A proportional-integral-derivative (PID) controller is used to actively lock this derivative signal to be maintained at zero value, and a compensatory signal is fed back to the bias circuit of the QCL. In this way, the QCL frequency can be stabilized to a particular molecular absorption line.

We measured the methanol absorption spectrum (versus frequency) by sweeping the QCL bias voltage, since the voltage regulates its emission frequency. Fig. 2 shows an obtained strong absorption line at around 3.45 THz. Also shown in the figure is the first derivative of the absorption profile, which is obtained by using a lock-in technique. The lock-in derivative signal is in turn utilized for the PID control loop, whereby a feedback signal is generated and fed to the bias circuit of the QCL in order to yield a stabilized lock-in signal. Around the frequency of the absorption peak, the linear region of the lock-in signal indicates the frequency locking range. We apply the exact same locking scheme for both QCLs. To illustrate the locking process, the inset of Fig. 2 plots the lock-in signal for laser A. When the laser is

unlocked, the observed fluctuations reflect the frequency fluctuations of the QCL. The observed 1 Hz frequency feature in the lock-in signal is the result of the typical temperature oscillation of the pulse tube cooler. After the PID feedback loop is enabled, the lock-in signal becomes well stabilized and maintained at the 0 mV setpoint. Thus, the laser frequency is stabilized.

The laser linewidth can be estimated by transforming the variation in voltage of the lock-in signal, which represents the error signal, into the frequency domain. The lock-in signal is first converted to the equivalent variation in the QCL bias voltage using the measured linear profile in the main figure of Fig. 2. Subsequently, two different methods are applied to calibrate the numerical correspondence between frequency and voltage. Method one is based on spectroscopic measurements,[12] where the frequency tuning coefficients of the QCL due to the bias voltage are obtained. Then, the frequency can be obtained by multiplying the voltage (the lock-in signal) with the tuning coefficient. The
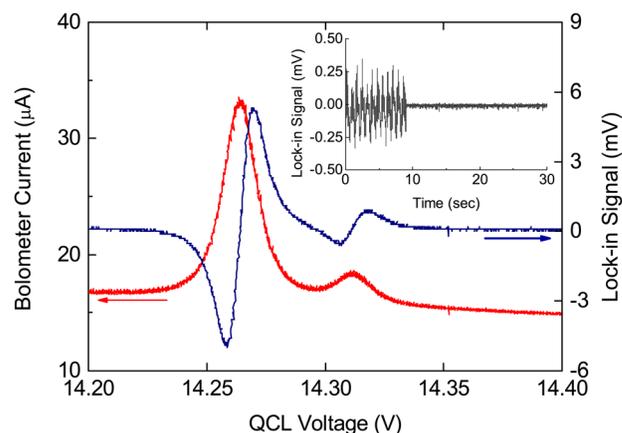
FIG. 2. (Color online) Absorption profile of methanol lines at 1.1 mbar as measured for "laser A." By using a NbN bolometer detector and a lock-in amplifier, the error signal was measured, labeled as lock-in signal. The voltage window (linear region for the lock-in signal) used for lock-in feedback loop is 8 mV. The inset shows the lock-in signal in the unlocked and locked state after about 8 s for "laser A."

second method is to make use of the methanol gas pressure broadening coefficient at 3.5 THz,[12] which is established from a set of methanol absorption lines at different gas pressures. In this way, a lock-in signal can also be transformed into frequency. We find that the two different approaches give similar results to within 10%. Fig. 3 plots a histogram of the relative frequency fluctuations of a 10 s observation time in order to present the standard deviation of the center freuqency. We find that the free running laser linewidth for laser A is roughly 1 MHz, while the locked linewidth is less than 18 kHz (both estimated using the FWHM of the profile). For laser B, we obtain the free running linewidth of around 3 MHz and the locked linewidth of 51 kHz, both of which are about 3 times larger than that found for laser A. We also notice that in Fig. 3 the free running line shape of laser B is double-peaked, while it is single-peaked for laser A. When locked, both QCLs show a Gaussian-like shape.

Now, we discuss the line shape and linewidth. It is known that the frequency noise of the QCL governs the free running line shape.[10] In our experiment, the frequency noise is mainly due to temperature and electric (DC and AC voltage signal) fluctuations. The temperature fluctuations are directly caused by the intrinsic fluctuations of the pulse tube cryocooler with a frequency around 1 Hz, while the electric fluctuations are due to the current noise in the DC bias and due to the AC modulation signal. The latter is typically at a relatively high frequency (~kHz).

We have simulated the line shape in this case by assuming that the temperature fluctuations, $A_T sin(2\pi f_T t)$, and AC voltage modulation, $A_E sin(2\pi f_E t)$, are the dominant fluctuations and that they give linear response to the frequency fluctuations, where $A_{T,E}$ and $f_{T,E}$ are the amplitude and frequency, respectively. We find that when the amplitude A_T and A_E are comparable, the line shape appears to be single peaked. In contrast, when $A_T \gg A_E$, the line shape becomes double-peaked. To verify this, we now examine what are the dominant fluctuations. For this, we measured the temperature and voltage tuning coefficients for each laser. We find that both lasers have a similar electric tuning coefficient, but a different temperature tuning coefficient. The latter for laser B is more than 2 times higher than laser A. Therefore, we conclude that the higher temperature tuning coefficient in laser B are predominant, which attributes to the observed double-peaked line shape. For the same reason, we also understand its larger free-running linewidth. As for why there is such a difference in the temperature tuning coefficient between two similar lasers, however, is unclear.

With regard to the locked linewidth, we notice that the obtained minimal linewidth is consistent with the published results in Ref. 4 where a linewidth of 30 kHz was measured within a short period of 3 ms, and in Ref. 14 where a a linewidth of 6.3 kHz was obtained with a different frequency locking technique.

Fig. 4 shows the measured noise power spectral densities of the lock-in signals for both lasers with and without the frequency locking. In this measurement, the 30-ms time constant set in the lock-in amplifier has induced a small noise bandwidth of ~5 Hz in the figure. The frequency locking results in suppression of the noise level over 30 dB at frequencies below about 5 Hz for both QCLs, illustrating the effective laser linewidth reduction. Furthermore, the higher frequency noise levels for laser B correspond to the measured larger linewidths. Although the noise data beyond 5 Hz may also be useful to understand the physical process, they are less straightforward because of the interplay between the lock-in bandwidth (~5 Hz) and the PID bandwidth (~1 kHz). This interplay determines the bandwidth of the entire feedback loop, which in turn governs the linewidth reduction factor.

In conclusion, we succeeded in frequency locking of two 3.5-THz DFB quantum cascade lasers by using methanol molecular absorption lines. Two different methods were
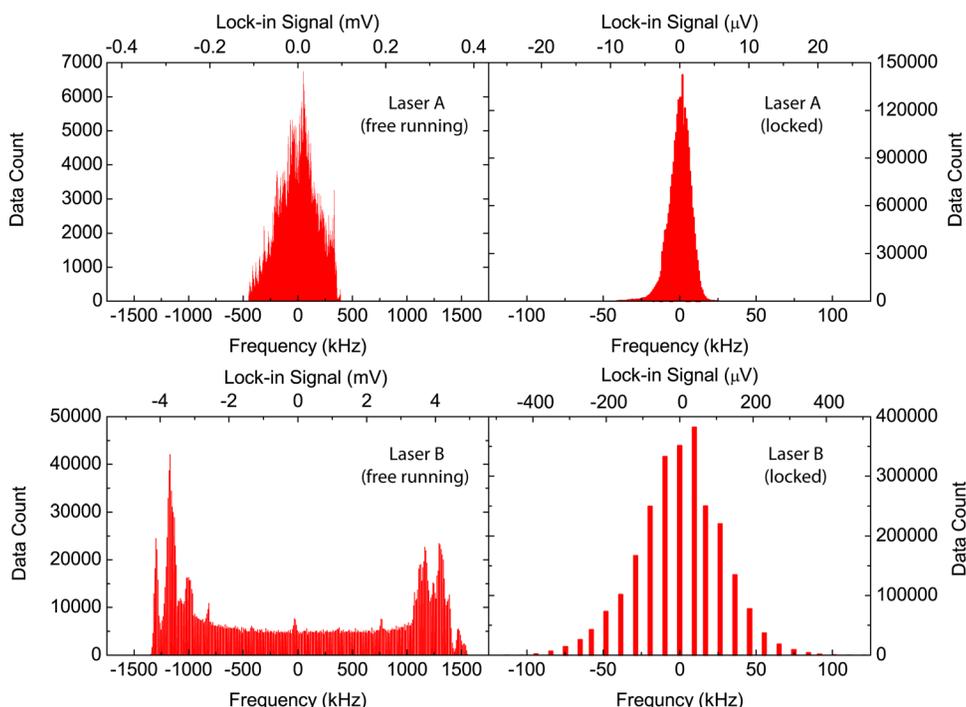
FIG. 3. (Color online) Histogram of the relative frequency deviation to the center frequency for two lasers in the free running and locked states, calculated based on the lock-in signal fluctuations of a 10 s observation time.
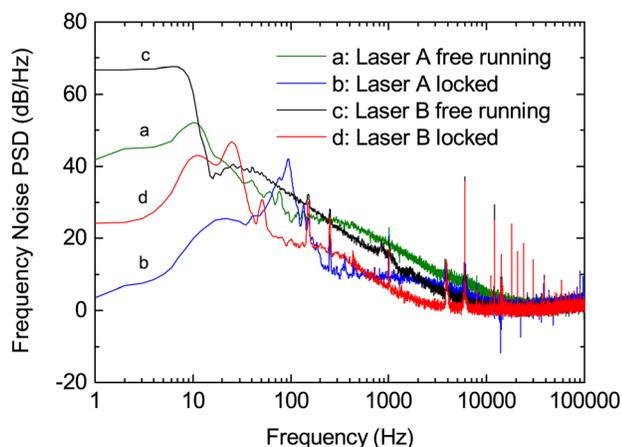
FIG. 4. (Color online) Frequency noise power spectral density of the lock-in signal for two lasers in the free running and locked states.

applied to convert the lock-in signal in voltage to the linewidth of the lasers, and both result in similar linewidths. The electrical and temperature tuning coefficients play a crucial role in determining the free-running line widths and shapes. The minimal linewidth is found to be below 18 kHz, which is sufficiently small for local oscillator applications. For the future practical applications like SOFIA, we plan to use a high-T_c (77 K and above) superconducting bolometric detector in place of the low temperature NbN bolometer shown here to simplify the instrument.

[1]B. S. Williams, Nat. Photonics **1**, 517 (2007).
[2]M. Yamanishi, T. Edamura, K. Fujita, N. Akikusa, and H. Kan, IEEE J. Quantum Electron. **44**, 12 (2008).
[3]C. Jirauschek, Opt. Express **18**, 25922 (2010).
[4]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 *et al.*, Opt. Lett. **29**, 575 (2004).
[5]S. Bartalini, M. S. Vitiello, L. Consolino, S. Borri, R. Eramo, A. Tredicucci, M. Inguscio, and P. D. Natale, in Proceedings of the 11th International Conference on Intersubband Transitions in Quantum Wells, Badesi, 11-17 September 2011, "The intrinsic linewidth of 60 Hz was inferred from the measured frequency noise spectral density of a THz QCL" (unpublished).
[6]P. Khosropanah, A. Baryshev, W. Zhang, W. Jellema, J. N. Hovenier, J. R. Gao, T. M. Klapwijk, D. G. Paveliev, B. S. Williams, S. Kumar *et al.*, Opt. Lett. **34**, 2958 (2009).
[7]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).
[8]M. Ravaro, C. Manquest, C. Sirtori, S. Barbieri, G. Santarelli, K. Blary, J.-F. Lampin, S. P. Khanna, and E. H. Linfield, Opt. Lett. **36**, 3969 (2011).
[9]A. Wootten and A. R. Thompson, Proc. IEEE **97**, 1463 (2009).
[10]H. Richter, S. G. Pavlov, A. D. Semenov, L. Mahler, A. Tredicucci, H. E. Beere, D. A. Ritchie, and H.-W. Hübers, Appl. Phys. Lett. **96**, 071112 (2010).
[11]M. I. Amanti, M. Fischer, G. Scalari, M. Beck, and J. Faist, Nat. Photonics **3**, 586 (2009).
[12]Y. Ren, J. N. Hovenier, R. Higgins, J. R. Gao, T. M. Klapwijk, S. C. Shi, B. Klein, T.-Y. Kao, Q. Hu, and J. L. Reno, Appl. Phys. Lett. **98**, 231109 (2011).
[13]Y. Ren, W. Miao, Q. J. Yao, W. Zhang, and S. C. Shi, Chin. Phys. Lett. **28**, 010702 (2011).
[14]A. Baryshev, J. N. Hovenier, A. J. L. Adam, I. Kašalynas, J. R. Gao, T. O. Klaassen, B. S. Williams, S. Kumar, Q. Hu, and J. L. Reno, Appl. Phys. Lett. **89**, 031115 (2006).