Frequency stabilization of quantum-cascade lasers by use of optical cavities

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We report a heterodyne beat with a linewidth of 5.6 ± 0.6 Hz between two cavity-stabilized quantum-cascade lasers operating at 8.5 μm. We also present a technique for measuring this beat that avoids the need for extreme isolation of the optical cavities from the environment, that of employing a third servo loop with low bandwidth to force one cavity to track the slow drifts and low-frequency fluctuations of the other. Although it is not fully independent, this technique greatly facilitates heterodyne beat measurements for evaluating the performance of cavity-locked lasers above the bandwidth of the third loop.

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Quantum cascade lasers[1,2] (QCLs) emitting in the mid- to long-wave infrared have greatly increased the availability of tunable lasers in this spectral region. Applications for such laser sources include high-resolution spectroscopy,[3,4] optical communications,[5] and chemical sensing.[6] In this Letter we extend previous research on linewidth narrowing and frequency stabilization of QCLs, both free running[7] and locked to a molecular transition,[8] by locking these lasers to an optical cavity with the Pound–Drever–Hall technique.[9–11] Although they are semiconductor injection lasers by nature, QCLs have intrinsic linewidths that are expected to be much narrower than those of conventional near-infrared diode lasers operating at the same power. The reasons are that the photon energy and hence the Schawlow–Townes linewidth are smaller and that the alpha parameter is expected to be near zero,[1,12–14] which for diode lasers typically increases the limiting linewidths by factors of 10–40 above the Schawlow–Townes limit. In addition to the interesting physics of QCLs, their narrow infrared laser linewidths enable sub-Doppler spectroscopy, cavity-based ultrahigh-sensitivity absorption methods,[15–17] laser velocimetry, and vibrometry to be more easily performed.

Locking a QCL to an optical cavity requires servo control loops with minimal delay and sufficient bandwidth to control high-frequency fluctuations, and high gain at low frequencies to remove $1/f$ noise and drifts. One can measure the performance of such a servo loop by examining the noise at certain points within it. However, these measurements are by definition in-loop measurements, and inferring similar out-of-loop performance from them can lead to significant errors.[18] A more definitive indicator of potential performance is the linewidth of the heterodyne beat between two such frequency-stabilized lasers. This relative frequency measurement is typically taken while two lasers are locked to separate modes of a single optical cavity.[11,19] This mode-locked measurement relaxes some of the stringent engineering requirements that are necessary to make two fully independent optical cavities stable enough to facilitate the measurement.[20] In this Letter we present a technique that uses an extra servo loop, called a tracking servo, that allows us to use two separate optical cavities with even fewer engineering constraints. The cavities used in the experiment consisted of dielectric mirrors mounted with Viton O rings into stainless-steel vacuum fittings and were bolted directly to a standard optical table with only rudimentary vibration isolation. Intended as simple low-finesse cavity-enhanced sensors, these optical cavities were not constructed for ultrastable performance. With the tracking servo, we used information derived from the heterodyne beat itself to force one laser-cavity system to track the other at frequencies below the tracking servo unity gain bandwidth of 6 kHz. With only a 6 dB/octave roll-off, the tracking caused by this loop is significant only at frequencies approaching dc. Nevertheless, this tracking is sufficient to suppress the effects of considerable environmental disturbances to the optical cavities, allowing a highly stable 5.6 ± 0.6 Hz wide beat measurement to be taken. This value represents an effective reduction from the recently recorded free-running linewidth of the same lasers of 150 kHz (Ref. 7) of more than 25,000.

Figure 1 is a simplified schematic of the experiment. The two quantum-cascade lasers, QCL1 and QCL2, were fabricated at Bell Laboratories–Lucent Technologies, mounted in liquid-nitrogen-cooled Dewars, and powered by low-noise custom-built constant-current controllers used previously.[7] To facilitate the Pound–Drever–Hall locking we phase modulated the QCL drive currents at 15 and 9.93 MHz, respectively, to depths of 1.1 rad. The optical cavities, labeled CAVITY 1 and CAVITY 2 in Fig. 1, had free spectral ranges of 560 and 470 MHz and line widths of 2.72 and 2.67 MHz, respectively. A custom-built transimpedance preamplifier (Z) was individually

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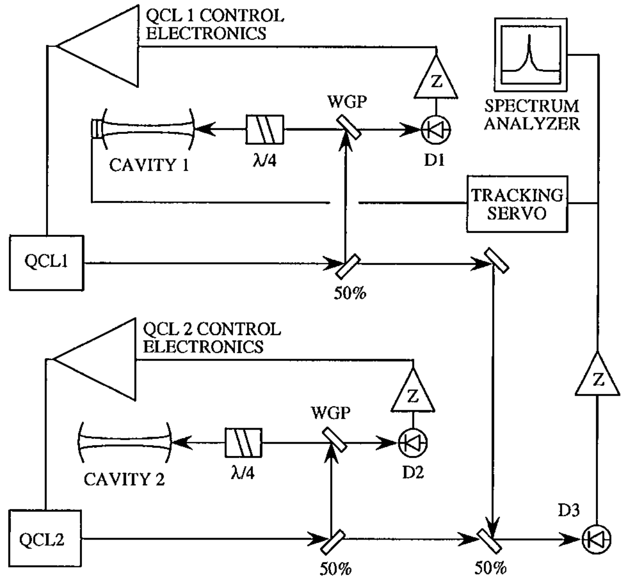

Fig. 1. Experimental diagram showing the beams from the QCLs reflecting from 50% beam splitters and being coupled into optical cavities by optical circulators made from wire-grid polarizers (WGPs) and quarter-wave rhombs ($\lambda/4$'s). The light reflected from the cavities is incident upon detectors D1 and D2. Light from the two cavity-locked QCLs is combined on a third 50% beam splitter and directed onto heterodyne beat detector D3, whose signal is monitored on either a swept spectrum of a fast-Fourier-transform spectrum analyzer. When the tracking servo is active, it removes low-frequency differential noise by acting on the length of cavity 1 by means of a piezoelectric element mounted on one mirror.

matched to each of the liquid-nitrogen-cooled mercury cadmium telluride detectors, D1–D3. The detector preamplifier combinations were shown to have input current noise values of ~4 pA/$\sqrt{\text{Hz}}$. This value was approximately equal to the shot-noise contribution in each case; the dc photocurrents were in the range 50–65 μA. The 100-μm-diameter heterodyne detector permitted operation to beyond 600 MHz. The unity gain frequencies of the laser-cavity locking servo units were measured to be beyond 1.5 MHz, and the two proportional-integrator stages in each unit provided very large gains at low frequency, measured to be greater than 10^6 at 10 Hz.

Initial experiments used a rf spectrum analyzer and no tracking servo. For subsequent experiments we mixed the beat signal from typically 40 MHz to ~50 kHz, where we used a fast-Fourier-transform audio spectrum analyzer (SR785) that has much finer resolution and whose time-domain sampling simultaneously measures all frequencies within its passband. For the highest-resolution measurements we employed the tracking servo to minimize the relative frequency changes between the two cavities. Some of the heterodyne beat signal was passed to a tracking filter,[21] which uses a double-balanced mixer to phase lock a voltage-controlled oscillator to the heterodyne beat. The voltage-controlled oscillator's control voltage, which tracks the heterodyne beat frequency, was amplified, integrated, and applied to the piezo element in cavity 1; it greatly suppressed low-frequency drift and acoustic noise. However, the optical table was still floated on air legs, and the experiment was performed in a quiet environment.

Figure 2(a) shows a linear scale heterodyne beat spectrum taken with the tracking servo active. The span is 15 kHz, and the resolution bandwidth (RBW) is 32 Hz. The central feature is the principal beat. The series of lobes roughly 2 kHz apart to either side of this central feature is due to excited mechanical resonances in the mirror mounts in both optical cavities, identified by use of loudspeaker for local acoustic excitation.

Figure 2(b) shows another beat spectrum but recorded with a much narrower span to show only the central feature and also with a much reduced RBW of 2 Hz. Because the filter of the analyzer can be approximated as Gaussian, and the lineshape that is due to small fast frequency fluctuations over a large bandwidth is Lorentzian,[22] we used a Voigt fit to extract the width of the heterodyne beat. This fit has a Gaussian part of 2.0 ± 1.2 Hz, corresponding to the analyzer's RBW, and a Lorentzian part of 5.6 ± 0.6 Hz, corresponding to the relative beat linewidth of the two stabilized lasers.

Other measurements that we took without using the tracking servo showed beat linewidths of less than 1 kHz that were dominated by relative drifts and vibrations of the two cavities for measurement times longer than 300 ms. With the tracking servo active, these effects are strongly suppressed. It must be recognized, however, that the linewidth of the beat signal under these conditions is not an entirely independent measurement of relative frequency stability. Such a measurement would require extreme attention to the reference cavities themselves and also to the surrounding environment, as discussed in the literature,[11,19,20] and providing this attention was not our intention. Rather, we wished to demonstrate the high level of performance of the stabilized QCL systems that we have developed. Contributions to the heterodyne beat linewidth from frequency fluctuations above the unity gain bandwidth of the tracking servo, 6 kHz, are independent. Hence we can infer that our measurement is an upper limit to the linewidth that will result from the small and fast frequency fluctuations above

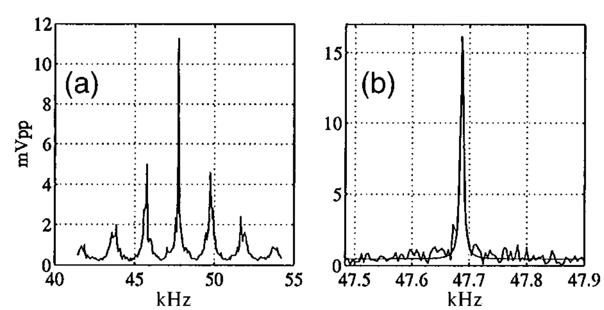

Fig. 2. Heterodyne spectra of two cavity-locked quantum-cascade laser systems: (a) shows the principal beat and 2-kHz-spaced acoustic noise structures and (b) was taken with a smaller span and RBW to show only the central feature and a Voigt fit to this feature.

6 kHz that are not removed by the main cavity-locking servos.[22]

There are several things to consider about the action of the tracking servo at frequencies below its unity gain bandwidth. Because the servo's gain is inferior to those of the principal servos, it could never enhance their absolute suppression capabilities. The tracking servo could, however, remove relative fluctuations that result from drifts or noise in the main servo reference voltages, or shifts in the zero point of the Pound–Drever–Hall error signal that were due to residual amplitude modulation. Consequently this measurement cannot attest to the purity of such references or to immunity to the effects of residual amplitude modulation.

As the RBW of any beat measurement is reduced, frequency fluctuations become more exposed not only as a result of the increased resolving power of a narrower window but also because a decrease in RBW results in a proportionate increase in acquisition time. Consequently there is an optimum RBW, which we found to be 2 Hz for the measurements presented here, above which the linewidth is dominated by the RBW and below which the slow drifts and frequency fluctuations dominate the spectra because of the long measurement times. In our case, small fluctuations in the tracking filter lock (possibly resulting from optical amplifier offset drifts, voltage-controlled oscillator frequency drifts, or finite integrator loop gain) resulted in small slow drifts in the error signal and hence in the heterodyne beat frequency that was being measured. Consequently the linewidth obtained is only an upper limit to the possible attainable cavity-locked laser performance. Indeed, for our experimental conditions, theoretical calculations similar to other found in the literature[19] deduced from cavity linewidth and detector noise yield a minimum beat line width of 20 mHz.

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