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Narrow-line phase-locked quantum cascade laser in the 9.2 micron range.

Franck Bielsa,^{1,2} Albane Douillet,^{1,2} Tristan Valenzuela,^{1,2} Jean-Philippe Karr,^{1,2,*} and Laurent Hilico^{1,2,*}

¹Laboratoire Kastler Brossel, Université Pierre et Marie Curie

T12, Case 74, 4 place Jussieu, 75252 Paris, France

²Département de Physique et Modélisation, Université d'Evry Val d'Essonne

Boulevard F. Mitterrand, 91025 Evry cedex

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We report on the operation of a 50 mW continuous wave quantum cascade laser (QCL) in the 9.2 μm range, phase locked to a single mode CO₂ laser with a tunable frequency offset. The wide free-running emission spectrum of the QCL (3-5 MHz) is strongly narrowed down to the kHz range making it suitable for high resolution molecular spectroscopy.

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Continuous wave high power (≥ 50 mW) quantum cascade laser sources (QCL) recently became commercially available [1]. They exhibit new features for infrared laser spectroscopy. Mid-IR QCLs are easily and widely tunable over more than 200 GHz by adjusting their temperature or injection current. Since they do not present phase-amplitude coupling, their ultimate linewidth is expected to be very narrow. In practice, due to thermal instabilities, they present a wide free-running emission spectrum, in the MHz range [2, 3]. Several experiments have shown a significant reduction of the QCL linewidth down to the 10 kHz range by injection current locking to a molecular line [4], or well below the kHz range by locking to a Fabry Perot cavity [5] using the Pound-Drever-Hall technique.

Phase-locking is a well-known technique used to reduce the relative phase noise between two laser oscillators or to transfer the spectral features of a stable laser to a noisy one [6]. This method consists in comparing the phase of the laser beat note with that of a RF synthesizer signal. It has recently been applied to the frequency control of terahertz QCLs [7], but never so far to infrared QCL linewidth reduction.

Our motivation for developing a frequency controlled QCL source is high resolution vibrational spectroscopy of the hydrogen molecular ions H₂⁺ or HD⁺. Indeed, those ions have recently been pointed out as good candidates for optical proton to electron mass ratio determination [8, 9, 10, 11]. Recent calculations have shown that two-photon vibrational spectroscopy of H₂⁺ in the 9.1-9.3 μm range is feasible [8] with interaction times of a few tens of ms. Transitions frequencies have been predicted with 1 ppb relative accuracy. Further progress in QED correction calculations should allow proton to electron mass ratio determination with a relative accuracy of 10⁻¹⁰, in significant progress over the present one (CODATA 2002) [14]. This corresponds to an uncertainty of a few kHz on the transition frequencies, hence the need for a kHz linewidth laser source.

In the 9.1-9.3 μm wavelength range, only two kinds of cw laser sources are available : CO₂ lasers and QCLs. Single mode CO₂ lasers can deliver up to several watt of optical power in a narrow bandwidth of less than 1 kHz. Unfortunately, their tunability covers a range of a few tens of MHz and the CO₂ emission lines do not overlap with the H₂⁺ spectrum. In this paper, we show that it is possible to combine the advantages of both kinds of sources by transferring the spectral properties of a CO₂ laser to a widely tunable QCL using phase-lock loop techniques. We demonstrate the operation of a tunable narrow-line high-power laser source suitable for two-photon spectroscopy of hydrogen molecular ions with a kHz resolution.

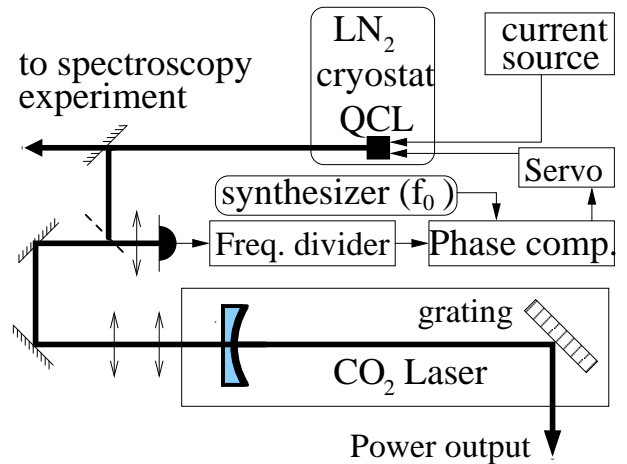


FIG. 1: Simplified experimental setup. The low power CO₂ laser output (≈ 50 mW) is beaten with a fraction of the QCL emission (≈ 10 mW). The frequency divider and phase comparator are described in text, and the servo circuit in Fig. 2.

The experimental setup is depicted in Fig. 1. The quantum cascade laser (SBCW496DN from AlpesLaser) is a single mode distributed feedback laser. When operated in cw regime under cryogenic conditions, it delivers up to 160 mW and is tunable between 9.16 and 9.24 μm (32.44 and 32.73 THz). Its threshold current is 400 mA at 77 K, and its maximum driving current is 1 A. The

*Electronic address: hilico@spectro.jussieu.fr

temperature and current tunabilities are 3 GHz/K and 150 MHz/mA.

The QCL is mounted in a liquid nitrogen cryostat with a ZnSe output window and driven by a home-made, low-noise stabilized current source. The voltage across the QCL is about 9 V and the dynamic resistance is 1.8 Ω . The electrical power dissipated in the QCL can be as high as 9 W, which requires efficient heat dissipation. For this purpose, the laser chip is fixed on a monolithic copper post, screwed on the cryostat cold plate in order to minimize the thermal resistance. The post can be heated and temperature controlled at the 10 mK level. With a 700 mA driving current, the QCL temperature stabilizes around 80 K without external heating and the QCL delivers an optical power of 50 mW at 9.166 μm , the required wavelength to probe the $(v=0, L=2) \rightarrow (v=1, L=2)$ two-photon line in H_2^+ .

The output beam is collimated using ZnSe collimating optics from Cascade Technologies (CO-01, 0.85 mm N.A. and 1.6 mm working distance). The far field transverse structure of the QCL beam presents a nodal structure with one main and two secondary lobes along the vertical axis. Along the horizontal axis, the Gaussian beam shape dependence on propagation distance is consistent with a M^2 parameter of about 3.3.

We use the CO_2 laser both as local oscillator to characterize the QCL spectrum properties, and as stability reference to phase lock the QCL. It is a sealed-off, low pressure, dual-discharge-arm, 1 m long single longitudinal mode laser. The cavity is closed by a R_{max} mirror at 9.2 μm and a 150 lines/mm grating in the Littrow configuration. The grating $zero^{th}$ diffraction order is the main output of the laser (95% efficiency). When operated with 13 Torr of standard gas mixture ($\text{CO}_2\text{-He-N}_2$) and 24 mA discharge current, the laser oscillation is obtained in the 9 μm band up to the 9R(48) line with more than 1 W of optical power. The CO_2 laser emission spectrum has a linewidth in the kHz range [15] and only very slow drifts (less than 1 MHz/s).

About 10 mW of QCL and 50 mW of CO_2 laser optical powers are mixed on a room temperature HgCdZnTe fast photodetector followed by a 37 dB low-noise RF amplifier. Although the beams' overlap is rather low, we obtain a signal to noise ratio of more than 45 dB in a 1 MHz resolution bandwidth for a beat frequency f_0 up to 1.5 GHz. The free-running QCL emission spectrum (Fig. 3a) is about 3 to 5 MHz wide as already observed with other QCLs [2, 3] and exhibits a low frequency jitter over more than 10 MHz.

In order to efficiently phase lock the QCL on the CO_2 laser, we tailor a wideband phase error signal and use a standard second order feedback loop [6, 16, 17]. The optical and electronic paths are shortened as much as possible (about 2 m) to minimize time delay. The beat note signal is high-pass filtered above 700 MHz and divided by 8 using a MC12093 high speed frequency divider. The phase comparison with a synthesized reference signal is performed at $f_0/8$ using a MCH12140 phase/frequency

detector with a $\pm 2\pi$ range. As a result, we obtain a very wide band (≥ 10 MHz) $\pm 16\pi$ phase/frequency comparison of the two lasers' spectra. The measured slope of the error signal is $s = 0.022$ V/rad.

The servo loop is depicted in Fig 2. It simply consists in an integrator with a 600 kHz cut-off frequency built with a fast operational amplifier. The feedback loop gain is adjusted using a variable resistor. The correction signal is directly applied to the QCL through a $\approx 1\text{k}\Omega$ resistor that limits the output current from the amplifier. This resistor is split into two parts. The first one belongs to a phase advanced filter with 2.4 MHz cut-off frequency adjusted to optimize the loop bandwidth. The second part is directly soldered on the QCL pads inside the cryostat in order to minimize wire capacitance effects. Because we use a phase-frequency detector, we have to choose the suitable sign for the correction signal depending on whether the QCL frequency is red or blue detuned with respect to the CO_2 laser frequency. This is the aim of the second inverter follower amplifier.

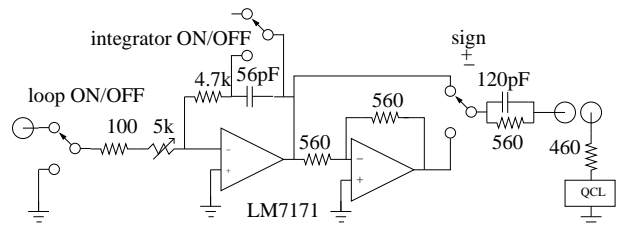


FIG. 2: Servo electronics of the phase lock loop. The resistor values are in Ω .

The beat note spectrum, taken with the tracking servo active, is shown in Fig. 3b. It represents the relative phase noise spectral density between the QCL and CO_2 laser. It exhibits an extremely narrow central peak with a -3 dB width of less than 200 Hz, limited by the resolution of the spectrum analyzer. This width is much smaller than the CO_2 laser one, meaning that the CO_2 laser's spectral features are transferred to the QCL. The spectrum's wings show a servo-loop unity gain frequency of the order of 6 MHz with a carrier 53 dB above the phase noise level in a 10 kHz resolution bandwidth. We now estimate the energy concentration ratio in the central peak. The normalized beat note spectrum can be expressed as [17]

$$S(f) \approx e^{-\sigma_\varphi^2} \delta(f) + S_\varphi(f) \quad (1)$$

where σ_φ is the phase error variance, $\delta(f)$ the Dirac delta-function and S_φ the phase noise spectral density. We have $S_\varphi(f) \approx 10^{-9.3} \approx 5 \cdot 10^{-10} \text{ rad}^2/\text{Hz}$. Graphical integration within the loop bandwidth gives an estimation of half the actual phase variance, i.e. $\frac{1}{2}\sigma_\varphi^2 = 3 \cdot 10^{-3} \text{ rad}^2$ [16, 17]. Fig. 4 shows the phase error spectral density at the output of the phase comparator. Comparison of curves (b) and (c) shows that the integrator reduces phase noise by more than 10 dB up to

200 kHz. The phase noise error signal spectral density exhibits a plateau at the $S_u=10^{-12.4}$ V²/Hz level corresponding to a phase noise spectral density $S_\varphi = S_u/s^2 = 8 \cdot 2 \cdot 10^{-10}$ rad²/Hz. Taking into account the 6 MHz noise bandwidth, we obtain an alternative estimation of the phase variance $\frac{1}{2}\sigma_\varphi^2 = 0 \cdot 005$ rad² in good agreement with the first one. From those values, we can deduce that the energy concentration ratio in the narrow central peak $e^{-\sigma_\varphi^2}$ is higher than 99%. Let us stress that the large feedback loop bandwidth is essential to obtain this result [18]. The phase locked operation of the QCL

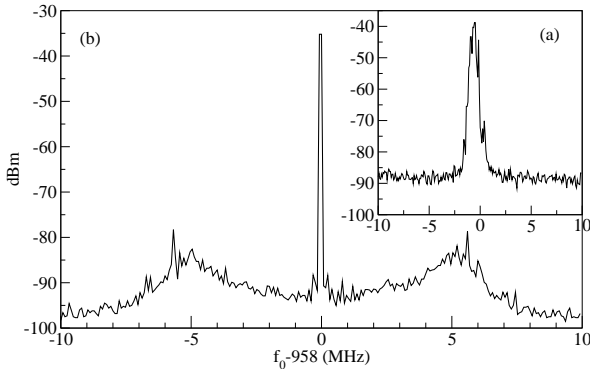


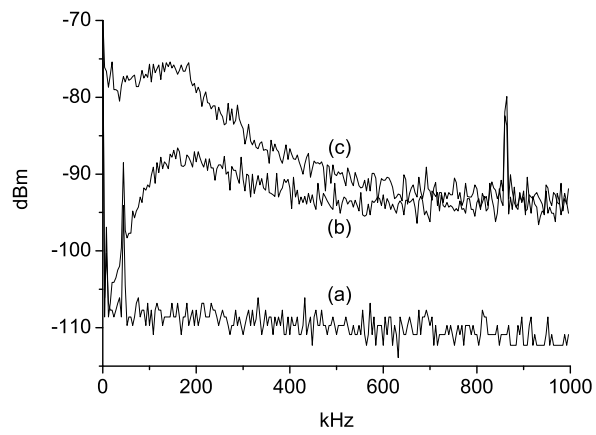
FIG. 3: (a) Free-running and (b) phase-locked QCL/CO₂ beat note spectrum. (a) RBW 500 kHz, VBW 1 kHz, (b) RBW 10 kHz, VBW 1 kHz.

is easily obtained with a tunable frequency offset in the 200-1500 MHz range around the CO₂ emission line. The high frequency limit is due to the detector cut-off. The low frequency limit is the minimum beat frequency necessary to obtain a wide band error signal after division and phase comparison. It can be overcome by frequency shifting the high power output of the CO₂ laser with an acousto-optic modulator before beating with the QCL. Stable operation of the lock during several hours is obtained.

In conclusion, we have shown that the spectral features of CO₂ lasers can be transferred to a QCL with a tunable frequency offset, making QCLs very interesting tools for high resolution molecular spectroscopy. We have used this source to observe several absorption lines of the ν_6 band of formic acid [19] in quasi-coincidence with the 9R36 to 9R42 emission lines of the CO₂ laser with a kHz resolution and measured their absolute frequencies [20] to obtain suitable molecular frequency references for H₂⁺ vibrational spectroscopy.

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FIG. 4: Phase noise spectral density of the QCL/CO₂ beat note. (a) electronic noise, (b) proportional and integral corrections, (c) proportional correction only. 1 kHz RBW and VBW.