Quantum cascade laser frequency stabilization at the sub-Hz level

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High-precision measurements with molecules may refine our knowledge of various fields of physics, from atmospheric and interstellar physics to the standard model or physics beyond it. Most of them can be cast as absorption frequency measurements, particularly in the mid-infrared ‘molecular fingerprint’ region, creating the need for narrow-linewidth lasers of well-controlled frequency. Quantum cascade lasers provide a wide spectral coverage anywhere in the mid-infrared, but show substantial free-running frequency fluctuations. Here, we demonstrate that the excellent stability and accuracy of an ultra-stable near-infrared laser, transferred from a metrological institute through a fibre link, can be copied to a quantum cascade laser using an optical frequency comb. The obtained relative stability and accuracy of $2 \times 10^{-15}$ and $10^{-14}$ exceed those demonstrated so far with quantum cascade lasers by almost two orders of magnitude. This set-up enables us to measure molecular absorption frequencies with state-of-the-art uncertainties, confirming its potential for ultra-high-precision spectroscopy.

Molecules are increasingly being used in precision tests of physics thanks to progress made in controlling molecular degrees of freedom12. They are now being used, for example, to test fundamental symmetries3–5 and to measure fundamental constants6–8 and their possible variation in time9–11. Most of these experiments are spectroscopic precision measurements and are often in the mid-infrared (MIR) domain where the molecules exhibit intense and narrow rovibrational transitions. This creates a need for efficient MIR laser sources, prompting efforts to develop ultra-stable and accurate continuous wave (c.w.) lasers as well as MIR frequency combs (refs 12 and 13 for instance). Quantum cascade lasers (QCLs) are promising c.w. sources—they are available anywhere in the 3–25 µm MIR range, and each QCL can be tuned over several hundreds of gigahertz. However, their free-running linewidth of tens to thousands of kilohertz makes their frequency stabilization challenging14–23.

Common references used for frequency stabilization in the MIR region include molecular rovibrational absorption lines3–26. However, molecular degrees of freedom cannot be controlled as efficiently as atomic ones, leading to limited frequency reproducibility and accuracy. Attempts to develop MIR ultra-stable cavities have been made, but their performances are far from those reported in the near-infrared (NIR) or visible regions27–28. It is thus appealing to use the best ultra-stable lasers as frequency references. As these are predominantly in the NIR region, it is necessary to bridge the gap between the NIR and MIR domains. This is possible using an optical frequency comb (OFC). The MIR frequency is locked to a high harmonic of the OFC repetition rate using sum- or difference-frequency generation processes. This not only provides ultimate stabilities of lasers locked to state-of-the-art ultra-stable cavities29, but also allows one to benefit from the direct link between such NIR sources and primary frequency standards30. A few groups have already demonstrated the stabilization of a MIR laser to a primary standard traceable frequency reference using an OFC17–19,21–23,31–33. Moreover, signals from a NIR ultra-stable laser referenced to a primary frequency standard can be transferred a few hundreds of kilometres over an optical-fibre link without any stability degradation34,35. Indeed, in the future, local reference lasers may no longer be required.

Following preliminary work using a CO2 laser32, we stabilized a QCL onto an OFC, itself locked to a remote NIR ultra-stable laser. The latter signal was transferred using a 43 km fibre link and its frequency monitored by primary standards. Because of the much larger free-running frequency noise of the QCL compared to the CO2 laser, the stabilization bandwidth had to be increased by three orders of magnitude. Evaluating the comb stability and the lock loops noise allowed us to demonstrate that the QCL frequency stability efficiently copies the comb stability. This gives almost two orders of magnitude improvement in the laser stability and linewidth compared with our previous work on a CO2 laser32 and previous works on QCLs21–24. It also results in a record frequency traceability. Furthermore, one main advantage of QCLs over CO2 lasers is their thousand times wider tuning range. By continuously tuning the stabilized QCL, we recorded OsO4 molecular absorption lines, some of them barely accessible with other sources. The obtained $8 \times 10^{-13}$ frequency uncertainty is well below any previously reported values for QCLs. This shows the potential of this set-up for precise spectroscopic measurements.

The experimental set-up is presented in Fig. 1. We used a room-temperature distributed-feedback QCL emitting up to 40 mW at 10.3 µm, with a tuning range of 60 nm (see Methods). This was phase-locked to an OFC consisting of an erbium-doped fibre mode-locked laser emitting around 1.55 µm. The comb repetition rate of $f_{\text{rep}} \approx 250$ MHz was stabilized by phase-locking one tooth of the OFC onto a NIR frequency reference generated at LNE-SYRTE laboratory and transferred to LPL laboratory through an optical-fibre link33 (see Methods). This NIR reference exhibits a relative frequency stability lower than $2 \times 10^{-15}$ between 1 and 100 s. Its absolute frequency is known with an uncertainty of $10^{-14}$ on this timescale when referenced to an H-maser via a local

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The comb repetition rate, \( f_{\text{rep}} \), is locked to the MIR reference (\( f_{\text{ref}} \)) by performing sum-frequency generation in an AgGaSe\(_2\) crystal (see Methods). The beat note of frequency \( \Delta_1 \) is processed to generate the error signal for the QCL phase-lock loop (PLL). The signal of frequency \( \Delta_2 \) on photodiode PD1 corresponds to the beat note between the MIR reference and the QCL. The stabilities and frequency noise \( \Delta_2 \) on photodiode PD2 corresponds to the beat note between the MIR reference and the QCL. The stabilities and frequency noise PSDs are evaluated by using a counter and a fast Fourier transform (FFT) analyser. Arrows indicate movable optics and the padlocks symbolize the contributions. The OFCs’ phase-lock loops. Ultra-stable laser #2 is used to evaluate the stability of the beat note between the MIR reference and the QCL (curve b, blue circles), the OFC (curve c, green triangles) and the beat note between the QCL and the CO\(_2\) laser, both stabilized onto the OFC (curve d, black stars). Overlapping Allan deviations are processed from data measured using a \( \Delta'_1 \) counter with \( 1 \) s gate time. For curve d, a dead-time free counter was used.

The performance of this set-up was assessed by measuring the relative frequency stability and frequency noise power spectral density (PSD) of the QCL. The stability of the QCL was first compared to that of a state-of-the-art secondary frequency standard around 10 \( \mu \)m, a CO\(_2\) laser stabilized on an OsO\(_4\) saturated absorber (26,36,37) (hereafter referred to as the ‘MIR reference’, see Methods). The beat note of frequency \( \Delta_1 \) between the stabilized QCL and this MIR reference was detected on photodiode PD2 (Fig. 1) and sent to a frequency counter. The stability of the beat note is the quadratic mean of the MIR reference and the QCL stabilities, and was obtained by calculating the overlapping Allan deviations of the data (red squares in Fig. 2). The fractional frequency stability is equal to 5 \( \times \) \( 10^{-17} \) for 1 s averaging time and decreases as \( \tau^{-1/2} \) up to a few tens of seconds. The stability of the beat note \( \Delta'_1 \) between the MIR reference and the OFC, detected on PD1, is also plotted for comparison (blue circles). The two stabilities are almost identical, except for small deviations due to non-stationary effects, and reflect the noise level of the MIR reference. Incidentally, this is the same noise level as has been measured previously\(^{12,36}\). The MIR reference contribution being dominant, this measurement can only provide an upper limit for the QCL stability.

The QCL frequency stability is expected to be given by the stability of the LPL OFC, with only a negligible contribution from the phase-lock loop residual frequency noise. Here, we evaluate both contributions. The OFC stability was assessed by beating an optical mode of the OFC with a second remote ultra-stable laser located at LNE-SYRTE (ultra-stable (US) laser #2, see Methods). The Allan deviation, shown as green triangles in Fig. 2,
The frequency noise PSD of the QCL phase-locked onto the NIR reference (QCL/OFC) and the MIR reference (curve a, red), the beat note between the MIR reference and the OFC (curve b, blue), the OFC (curve c, green), the noise-compensated 43 km optical-fibre link (curve d, brown), and the free-running QCL (curve e, black). All these PSDs are relative to a carrier frequency of 29.1 THz (10.3 µm wavelength).

As such, the link adds a minor contribution to the OFC stability and that both copy the OFC spectral properties. As shown in Fig. 2 (black stars), this is $2 \times 10^{-16}$ at 1 s averaging time, one order of magnitude below the OFC stability, and reaches $10^{-17}$ at 100 s. This demonstrates that the QCL and the CO$_2$ laser frequency fluctuations are identical at a level well below the OFC stability, and that both copy the OFC spectral properties. This stability is ten times better at 1 s than any other MIR laser to date$^{37}$, and constitutes an improvement by a factor of at least 30 over the previous record with a QCL$^{24}$.

To further characterize the QCL stabilization, the frequency noise PSD of the aforementioned OFC–ultra-stable laser #2 beat note signal was also measured. The result is presented in Fig. 3 (green curve c) after being scaled to 10.3 µm (see Methods). The frequency noise PSD is $10^3$ Hz$^2$ Hz$^{-1}$ at 100 kHz Fourier frequency (limited by the $\sim 500$ kHz locking bandwidth) and exhibits a plateau at $10^{-2}$ Hz$^2$ Hz$^{-1}$ between 1 and 100 Hz. Figure 3 also shows the frequency noise PSD of the 43 km noise-compensated fibre link$^{32}$ (brown line d). This is below or equal to the OFC PSD. As such, the link adds a minor contribution to the OFC noise. Figure 3 also displays the frequency noise PSDs of the beat notes at frequencies $\Delta_2$ between the MIR reference and the OFC-stabilized QCL (red curve a) and $\Delta_1$ between the MIR reference and the OFC (blue curve b). These two curves overlap almost perfectly below a Fourier frequency of 100 kHz. They are at a level of $10^2$ Hz$^2$ Hz$^{-1}$ from 1 Hz to 1 kHz and increase to $10^3$ Hz$^2$ Hz$^{-1}$ at 100 kHz. This noise level is the sum of the noise contributions from each laser. At low Fourier frequencies (below 1 kHz), the MIR reference noise dominates over the OFC noise, as previously measured in ref. 32. The bump around 400 Hz comes from the MIR reference locking bandwidth. Above 1 kHz, PSDs a and b overlap with the OFC noise PSD. Finally, we note a bump around 500 kHz on PSD a, corresponding to the QCL locking bandwidth. These measurements confirm that the QCL noise is indeed copying the OFC noise. Figure 3 also displays the free-running QCL PSD (black curve e), highlighting a 12 orders of magnitude reduction in the QCL frequency noise at 1 Hz.

The relevant parameter for spectroscopic applications is the emission linewidth, which we evaluated from the measured OFC frequency noise PSD following the approach in ref. 38. The low-frequency contribution needed for this calculation was deduced from the 1 s gate time frequency data used for deriving the OFC stability of Fig. 2. The resulting QCL line shape shown in Fig. 4a exhibits a full-width at half-maximum (FWHM) of 0.2 Hz (for a 10 mHz resolution bandwidth), which is the narrowest linewidth reported for a QCL by far. It can be compared to the calculated free-running line-width of 300 kHz (found using a 1 kHz resolution bandwidth). Figure 4b shows the beat note signal $\Delta_2$ between the phase-locked QCL and the MIR reference, recorded with a resolution of 125 mHz. A Lorentzian fit gives a 10 Hz FWHM. Although dominated by the MIR reference contribution, such a narrow beat note has never before been recorded with a QCL.

The frequency accuracy, or more specifically the frequency traceability to the primary standards of LNE-SYRTE$^{39}$, was ensured by using phase-lock loops for QCL frequency stabilization. The QCL absolute frequency is thus known with an uncertainty of $\sim 10^{-14}$ after 100 s averaging time, when the NIR laser is referenced to the H-maser only. By averaging for long enough, the $3 \times 10^{-16}$ Cs fountain accuracy$^{30}$ can ultimately be transferred to the NIR reference and in turn to the QCL frequency.

Such an accurate and ultra-stable QCL is ideal for carrying out molecular spectroscopy at the highest resolutions in the MIR spectral region. As a first demonstration we performed saturated absorption spectroscopy of the OsO$_4$ molecule at an unprecedented resolution and accuracy for QCL sources. OsO$_4$ is used as a test molecule for high-resolution frequency measurements in the MIR region as many tens of its lines have been accurately measured to serve as secondary frequency standards. The QCL beam was sent through an OsO$_4$-filled Fabry–Perot (FP) cavity. Saturated absorption spectra were recorded in transmission through this cavity by scanning the RF oscillator used to phase-lock the QCL onto the OFC (see Methods). This allowed the laser to keep its extremely high spectral purity, frequency stability and accuracy while being swept.

Figure 5 presents two OsO$_4$ lines in a spectrum spanning 5 MHz in the vicinity of the CO$_2$ R(14) 10 µm emission line. The QCL beam was frequency-modulated at 9.5 kHz and third harmonic detection was used. The line on the left is the unidentified (that is, not assigned unequivocally to a molecular transition) $^{185}$OsO$_4$ reference line in the vicinity of the CO$_2$ R(14) 10 µm emission line.

This estimation is based on the temporal data used to derive the OFC relative stability (curve c of Fig. 2) and the OFC frequency noise PSD (curve c of Fig. 3). The beat note between the QCL phase-locked onto the MIR reference and the MIR reference, recorded with a FFT analyser (125 mHz resolution, average of 10 sweeps of 8 s). The beat note is offset to zero by subtracting $\sim 50$ kHz. The red line is a Lorentzian fit of the data with a linewidth (FWHM) of 10 Hz.
the OsO₄ absolute frequency grid. Its frequency is reported to be ν₃₉ = 29,137,747,033,333 THz, reported in ref. 39. To our knowledge, this is the first time the other line (on the right) has been measured. The inset in Fig. 5 is a 200 kHz span spectrum as typically used to determine line centre absolute frequencies. It shows a signal-to-noise ratio (SNR) of about 200 for a total recording time of 400 s and peak-to-peak linewidth of 25 kHz. The data are fitted to a combination of a third and a fifth derivative of a Lorentzian.

Exhaustive studies of systematic effects affecting OsO₄ line shifts have already been carried out. In this work, care was taken to set experimental conditions similar to those reported in the literature for the reference line, to allow the comparison of absolute frequencies. We evaluated the frequency of a given line as the weighted mean frequency of independent measurements (see Methods). The 190OsO₄ reference line frequency was found to be ν₃₉ = 9 Hz, with a standard uncertainty of the mean of 22 Hz. The frequency of the unreported line shown in Fig. 5 was measured to be 4,147,399(23) Hz. The absolute frequencies of three other lines were measured and are listed in Table 1. Our results, which are in very good agreement with those reported in the literature, lead to fractional uncertainties on absolute frequencies as low as 8 x 10⁻¹³. This is one of the lowest uncertainties ever reported using saturated absorption spectroscopy in the MIR region. In particular, the uncertainty of the reference line is limited by the choice of frequency modulaton parameter. This simple model allows us to extract the absolute frequency of the line centre with a typical 50 Hz uncertainty given by the nonlinear regression, of the order of the standard deviation of independent measurements (47 Hz, for instance, for six measurements of the reference line).

Figure 5 | OsO₄ spectrum, in the vicinity of the R(14) emission line of CO₂.

Two lines are recorded over a span of 5 MHz in steps of 5 kHz, using third harmonic detection. The x axis is offset by ν₃₉ = 29,137,747,033,333 Hz, corresponding to the reported absolute frequency of the OsO₄ reference line (left-hand line). The right-hand line has not yet been reported in the literature. Inset: spectrum of the reference line recorded over 200 kHz in steps of 1 kHz. These data are fitted to the sum of a third and fifth derivative of a Lorentzian.

Table 1 | Absolute frequencies of five OsO₄ absorption lines in the vicinity of the R(14) CO₂ laser line.

| OsO₄ lines in the vicinity of the CO₂ R(14) laser line at 10.3 µm | Frequency shift from ν₃₉ calculated from refs 39 and 41 (kHz) | Frequency shift from ν₃₉ measured in this work (kHz) |
|---------------------------------------------------------------|----------------------------------------------------------|---------------------------------------------------|
| 190OsO₄ reference line (unassigned)                          | 0.000 (40)                                               | −0.009 (22)                                      |
| Unreported line                                              | −                                                      | +4,147,399 (23)                                   |
| 190OsO₄, R(46)A₁(−)                                         | +101,726.83 (5)                                          | +101,726.821 (32)                                 |
| Unreported line                                              | −                                                      | +123,467.401 (32)                                 |
| Unreported line                                              | −                                                      | +204,269.162 (33)                                 |

The frequencies are given with respect to the OsO₄/CO₂ R(14) reference line frequency, ν₃₉ = 29,137,747,033,333 THz, reported in ref. 39. In the second column we report the absolute frequencies calculated from refs 39 and 41 with 1σ uncertainty. The third column displays the results of this work, where the uncertainty is the standard uncertainty of the mean. The R(46)A₁(−) line has previously been recorded at lower pressure. Our measurement is thus expected to be pressure-shifted by approximately +10 Hz (ref. 26).
fundamental vibration of H2, but the laser frequency control is a serious experimental limitation.13 The electron-to-proton mass ratio and its possible temporal variation can be measured by high-resolution molecular spectroscopy, but this requires sources of the highest accuracy. Ultra-stable QCLs are also needed to perform precision spectroscopic measurements of molecules of interest for atmospheric studies, such as ozone or water, and preferentially in the 8–14 μm window.23,46. At LPL, this QCL-based spectrometer is critical for our ongoing efforts to make the first observation of parity violation induced by chirality at the 8–14 μm atmospheric window.49 In this project, vibrational frequencies of two enantiomers are measured and compared. The expected frequency difference is small, predicted to be on the order of 10−13 at most. Naturally, the candidate molecules have resonances in different spectral regions, many of which only QCLs can attain. This particular combination of precision, stability and flexibility of tuning in the MIR requires a very particular laser set-up, of which the first proof-of-principle is successfully demonstrated in this Article.

Methods

Methods and any associated references are available in the online version of the paper.

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Methods

The QCL. We used a commercial device from Alpes Laser. This can be tuned from 967 to 973 cm⁻¹ (corresponding to a tuning range of 40 nm or 180 GHz) by varying the temperature (between 283 K and 243 K) and the driving current. Cooling was provided by a chilled water circuit and a Peltier module driven by a commercial temperature controller. The threshold current is 570 mA. The QCL can be operated up to 870 mA and exhibits a maximum output power of 40 mW. It was driven by a homemade low-noise current source, resulting in a negligible contribution of the current driver to the free-running frequency noise (below $10^{-10}$ Hz² Hz⁻¹, down to 1 kHz). The frequency noise of the free-running QCL is shown in Fig. 3 (black curve e). This is $10^{-10}$ Hz² Hz⁻¹ at 1 Hz and has a slope of approximately 1/2 up to 250 kHz, followed by a steeper slope for higher Fourier frequencies.

LPL OFC stabilization to a NIR reference. The NIR frequency reference signal was generated at LNE-SYRTE laboratory. This signal was provided by a 1.54 µm c.w. laser with its frequency locked onto an ultra-stable cavity by the Pound–Drever–Hall method. This laser (AgGaSe₂, Dalin Optics) was controlled by the SYRTE-OFC measurement system to remove the slow frequency drift of the ultra-stable cavity. This gave an ultra-stable NIR frequency reference traceable to primary standards (H-maser) with a $10^{-12}$ uncertainty after 100 s. This reference signal, with frequency $\nu_{1}$, was then transmitted to LPL through a 43-km-long optical link (see above) and a correction of the light propagation in the optical fibre was implemented. The residual phase noise instability added by the compensated link (detected with full bandwidth) was below $10^{-13}$ at 1 s averaging time and decreased with a slope of $\nu^{-1}$ (ref. 32). This is below the stability of the ultra-stable laser. The signal of the ultra-stable-frequency reference was transferred to LPL without degradation (up to Fourier frequencies of a few tens of hertz). At LPL, it was used to phase-lock a local 1.54 µm laser diode for signal regeneration. The repetition rate $fi_{rep}$ of the LPL OFC was in turn phase-locked to this laser diode after removal of the carrier-envelope-offset frequency $f_{0}$ (ref. 47). Note that for this lock we used some small fraction of light at 1.55 µm present in the output beam at 1.82 µm (see section ‘QCL stabilization of the OFC’)48. Fast and slow corrections were applied, respectively, to an intra-cavity electro-optic modulator and a piezo-electric transducer (PZT) acting on the cavity length, resulting in a bandwidth of more than 500 kHz. The beam was then operating in the ‘narrow-linewidth regime’49.

QCL stabilization onto the OFC. The 1.55 µm LPL OFC was used to transfer the spectral properties of the NIR reference to the QCL. An additional output of the comb centred at 1.82 µm (total power 25 mW, mode frequencies $\nu_{OFCS}$, $\nu_{OFCD}$, where $\nu$ is an integer) was overlapped with the QCL beam (5 mW, frequency $\nu_{QCL}$) in an AgGaSe₂ crystal to perform sum-frequency generation. The polarizations of each beam were adjusted to be parallel. A set of lenses was used to focus the QCL beam in the crystal. An iris diaphragm and an optical isolator, composed of a wire-grid polarizer and a quartz optical isolator, minimized the optical feedback to the cavity (mainly caused by back-reflection from the AgGaSe₂ crystal facets52). Sum-frequency generation between the 1.82 µm comb and the 10.3 µm c.w. beam resulted in a shifted comb (30 mW, mode frequencies $\nu_{OFCS} + \nu_{OFCD} + \nu_{QCL}$) at 1.55 µm. This shifted comb was then combined with the original 1.55 µm output of the LPL OFC (10 mW, mode frequencies $\nu_{OFCS}$, $\nu_{OFCD}$) and a carrier-wave frequency $\nu_{rep}$, resulting in the carrier-envelope-offset-free frequency $\nu_{OFCS}$ with a much better signal-to-noise ratio (SNR). The beat note at frequency $\nu_{rep}$ was thus used as the local oscillator for all subsequent measurements. The resulting SNR was 9×10⁻⁴ at 1 s and decreased as $\nu^{-1}$, thus hardly contributing to the OFC stability53.

OFC frequency noise and stability. To obtain the LPL OFC spectral characteristics we evaluated the stability and frequency noise of the beat note between an optical mode of this comb and another ultra-stable laser (after removing the comb offset frequency $f_{0}$)54. This ultra-stable laser #2 consisted of a 1.54 µm c.w. source frequency locked onto an ultra-stable cavity similar to the one used to provide the NIR reference. The two ultra-stable lasers thus presented the same spectral properties. As they were transferred from LNE-SYRTE to LPL through the same optical-fibre link and because their frequencies were very close (only 0.375 MHz apart), the phase noise accumulated along the fibre link was approximately the same for both lasers. Thus, any propagation noise was cancelled in the beat note between the OFC and the second ultra-stable signal, and it was not necessary to compensate the link noise for this measurement. The beat note PSD (green curve c in Fig. 3) resulting from the sum of the OFC and the ultra-stable laser #2 noises thus gives an upper limit on the OFC frequency noise. The residual noise PSD of the stabilized fibre link (brown curve d in Fig. 3) should be added to it to obtain the overall frequency noise of the OFC. Nonetheless, the link noise contribution was found to be negligible except for Fourier frequencies between 20 and 400 Hz, where it becomes comparable to that of the OFC (leading, however, to a minor overall contribution). Note that these PSDs have been scaled to 10.3 µm by the square of the wavelengths ratio for a direct comparison with PSDs recorded at 10.3 µm. From the beat note frequency measurement, we also derived the OFC stability by calculating the Allan deviation $\sigma(\nu)$, assuming that the frequency noise $\nu$ and the Allan deviation $\sigma(\nu)$ are proportional. The correction applied to the beat note frequency was $\nu = (\nu_{OFCS} - \nu_{OFCD})/2$ (ref. 48). Fast and slow corrections were applied, respectively, to the intra-cavity electro-optic modulator and a piezo-electric transducer (PZT) acting on the cavity length, resulting in a bandwidth of more than 500 kHz. The beam was then operating in the ‘narrow-linewidth regime’49.

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