Frequency-comb-referenced quantum-cascade laser at 4.4 μm

S. Bartalini, P. Cancio, G. Giusfredi, D. Mazzotti, and P. De Natale

CNR-Istituto Nazionale di Ottica Applicata, Largo E. Fermi 6, 50125 Firenze FI, Italy, and European Laboratory for Non-linear Spectroscopy, Via Carrara 1, 50019 Sesto Fiorentino FI, Italy

S. Borri and I. Galli

Dipartimento di Fisica, Università di Firenze, Via Sansone 1, 50125 Firenze FI, Italy

T. Leveque

École Supérieure d'Optique, Institut d'Optique Théorique et Appliquée, 91403 Orsay Cedex, France

L. Gianfrani

Dipartimento di Scienze Ambientali, Seconda Università di Napoli, and CNISM-Unità Napoli 2, Via Vivaldi 43, 81100 Caserta CE, Italy

Received November 27, 2006; revised January 15, 2007; accepted January 25, 2007; posted January 30, 2007 (Doc. ID 77449); published March 19, 2007

We report what we believe to be the first absolute frequency measurement performed using a quantum-cascade laser (QCL) referenced to an optical frequency comb synthesizer (OFCS). A QCL at 4.43 μm has been used for producing near-infrared radiation at 858 nm by means of sum-frequency generation with a Nd:YAG source in a periodically poled lithium niobate nonlinear crystal. The absolute frequency of the QCL source has been measured by detecting the beat note between the sum frequency and a diode laser at the same wavelength, while both the Nd:YAG and the diode laser were referenced to the OFCS. Doppler-broadened line profiles of $^{13}CO_2$ molecular transitions have been recorded with such an absolute frequency reference. © 2007 Optical Society of America

OCIS codes: 120.3930, 190.2620, 140.3070, 140.3490, 300.6340, 300.6390.

In the past few years, optical frequency combs have proved to be a powerful metrological tool for the visible and near-infrared (NIR) region.[1] At present there is growing interest in extending the optical frequency comb synthesizer (OFCS) spectral coverage to the mid-infrared (MIR) region, which is currently lacking secondary frequency standards. Moreover, the fundamental vibrational transitions of most simple molecules (such as CO_2, CO, and CH_4) lie in the 2–12 μm spectral region: access to these wavelengths with low-noise and powerful radiation sources would allow unprecedented sensitivity in spectroscopic detection.

A first success in this direction has been obtained by difference-frequency generation (DFG) of MIR radiation starting from OFCS-referenced visible or NIR sources mixed in a nonlinear crystal.[2–4] The output power achievable by such nonlinear processes does not exceed the milliwatt range[5,6]: this can prevent quantum-noise-limited detection, thus limiting the achievable sensitivity of DFG-based spectrometers.

Quantum cascade lasers[7] (QCLs) operating in the MIR region represent a valid alternative to DFG systems. Thanks to their good stability, narrow emission linewidth and, above all, high emission power, these sources have been demonstrated to be extremely suitable for high-sensitivity and high-resolution spectroscopy,[8–11] with output powers as high as hundreds of milliwatts. The success in measuring the QCL absolute frequency would thus allow both high sensitivity and precision to be combined in the same MIR spectroscopic experiment.

In this Letter we report what is to our knowledge the first OFCS-referenced QCL working at 4.43 μm and its application to the observation of Doppler-broadened $^{13}CO_2$ spectral lines over an absolute frequency scale. For the metrological link to the OFCS, a frequency upconversion has been carried out by mixing a fraction of the QCL radiation with Nd:YAG laser emission at 1064 nm in a periodically poled lithium niobate (PPLN) nonlinear crystal. The sum-frequency generation (SFG) process provides a few microwatts of power at 858 nm, which produces a measurable radio-frequency signal when beaten on a fast photodiode with a diode laser working at the same wavelength. The frequency of the beat note can be easily counted, thus allowing an immediate measurement of the QCL frequency, provided that both the Nd:YAG and the diode laser frequencies (ν_Y and ν_D) have been referenced to the OFCS with phase-locked loops.

Our experimental setup is shown in Fig. 1. We have used a cw, liquid-N_2-cooled, distributed feedback (DFB) QCL (Alpes Lasers). Housed in a continuous-flow cryostat, it can be operated within the temperature range 80–100 K, with a stablity of a few millikelvins, as ensured by a proportional-integral-derivative controller. A homemade low-noise current driver (total current noise $<2.5\ \mu A_{RMS}$ in the 0–10 MHz band) powers the device (300 mA at 9 V). The QCL tunability has been characterized, obtaining 770 MHz/mA and 4.33 GHz/K for frequency tuning with current and temperature, respectively.

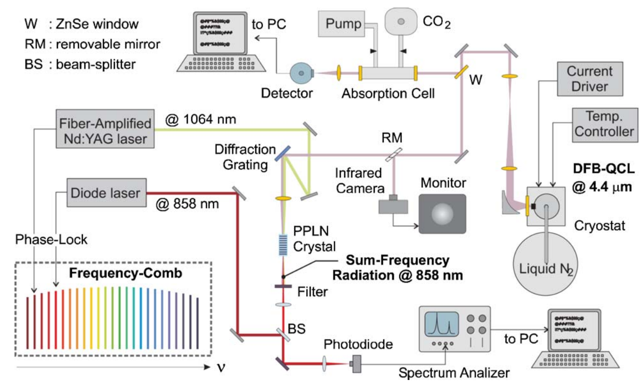

Fig. 1. (Color online) Schematic drawing of the experimental setup. PC, computer.

The strongly diverging QCL beam is collected and collimated by a short-focus paraboloidal reflector: the beam intensity profile is continuously monitored with a pyroelectric infrared camera. After being reduced in size by a telescope, the MIR radiation is split into two beams by a ZnSe window. The reflected beam is sent to a 15 cm long cell for CO_2 spectroscopy and is detected by a HgCdTe detector. The transmitted beam is superimposed on a fiber-amplified Nd:YAG beam by a diffraction grating and sent to a PPLN crystal for the SFG process. Both beams are focused into the crystal by the same 10 cm focal length CaF_2 lens. The choice of beam sizes and the lens focal length leads to a confocal parameter $b \simeq L$, where $L=4$ cm is the crystal length, for both beams. The optimal quasi-phase-matching condition in the crystal is reached for a poling period $\Lambda = 23.0\ \mu$m and temperature $T = 13\,°$C. With about 1.2 W of Nd:YAG power and only 2 mW of QCL radiation incident on the nonlinear crystal, about 10 μW of SFG radiation has been obtained. This is only about 1/8 of the theoretical power predicted for three-Gaussian-wave mixing[12]; such a discrepancy is not surprising, since the QCL beam greatly differs from a perfect Gaussian beam.

After the PPLN crystal, an interference filter at 858 nm transmits the SFG radiation, which is then collimated and superimposed on the beam of an external-cavity diode laser working at the same wavelength. A fast avalanche photodiode finally detects the resulting beat note (40 dB above the noise level when acquired with a 500 kHz resolution bandwidth).

The OFCS used in this experiment is a mode-locked femtosecond Ti:sapphire laser that operates in the 500–1100 nm region with a comb mode spacing of about 1 GHz.[4] This repetition rate is synthesized by a 10 MHz quartz oscillator disciplined by a GPS/Rb clock with a stability of about 6×10^{-13} in 1 s and an accuracy of about 1×10^{-12} in the worst case. These figures are transferred to the absolute frequencies ν_D and ν_Y by phase locking them to the nearest OFCS tooth at each wavelength.

For the Nd:YAG laser, a low bandwidth ($\simeq 500$ Hz) phase-locked loop is used to correct only the slow phase fluctuations present in this source, leaving its linewidth almost unchanged. For the diode laser a direct-digital synthesis technique was preferred,[13] which leads to an effective phase lock between the two laser sources through the OFCS, which acts as a transfer oscillator. The phase-noise contributions of the OFCS offset and repetition rate parameters are thus canceled out, and the diode linewidth narrows to the Nd:YAG one (a few tens of kilohertz), while their frequency difference remains constant. Calling $\Delta\nu_{S-D}$ the measured beat note between the sum frequency ($\nu_{\mathrm{QCL}} + \nu_Y$) and ν_D, the QCL absolute frequency is expressed as

$$\nu_{\mathrm{QCL}} = \nu_D - \nu_Y + \Delta\nu_{S-D}, \tag{1}$$

where care is taken to account for the correct sign of the beat signal. The difference between ν_D and ν_Y has in principle a null linewidth, so that most of the contributions to the beat-note linewidth and to its fluctuations originate from the QCL itself.

A real-time fast Fourier transform spectrum analyzer has been used for the acquisition of the beat-note spectrum over different time-scales. Figure 2 shows the Gaussian profile of the spectrum over 100 ms, with a linewidth of 1.9 MHz (HWHM), mostly due to the driver current noise contribution and probably to thermal fluctuations. The acquisition over 40 μs shows a narrower linewidth (1.4 MHz HWHM) as expected, since over this time scale the temperature fluctuations are uninfluential and the driver current noise contribution is reduced.[14,15] On a longer time scale, since the QCL is always working in the free-running condition, the frequency stability is also affected by even small temperature fluctuations, which induce a few megahertz of jitter.

Partitioning the QCL linewidth among the different sources of phase noise would be an important issue to be deeply analysed. However, this goes beyond the aim in this Letter and will be studied in the future.

The OFCS-referenced QCL has been used for a point-to-point acquisition of the $^{13}CO_2$ $(00^01-00^00)P(30)$ Doppler-broadened transition (see Fig. 3). Each point of the trace shown in the figure is the result of two simultaneous measurements: the amplitude of the absorption signal, digitized by a

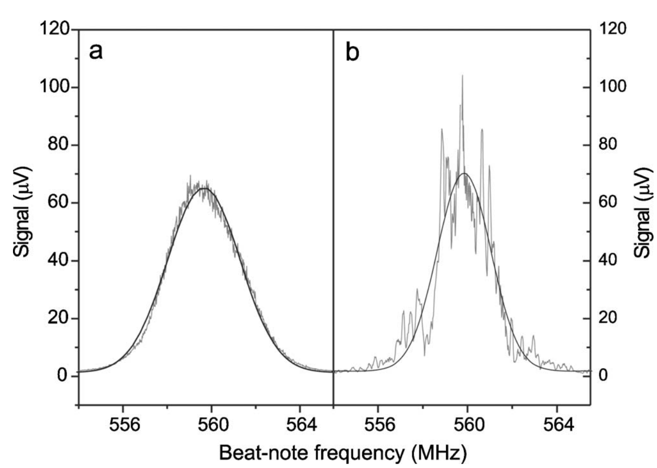

Fig. 2. Fast Fourier transform spectra of the beat note over time scales of, a, 100 ms and, b, 40 μs with their Gaussian fits.

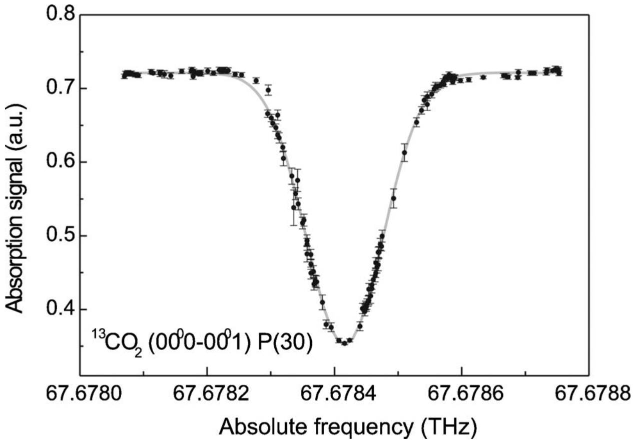

Fig. 3. Acquired $^{13}CO_2$ spectrum with $P = 33$ Pa. The nonuniform distribution of points is due to the slow temperature drift of the QCL during the acquisition. The light gray curve shows the Voigt fit of the data.

data acquisition board, and the peak frequency of the beat note, measured by a spectrum analyzer. A personal computer interfaced with both the data acquisition board and the spectrum analyzer has been used for synchronizing the acquisition and collecting the data. The frequency sweep across the line has been performed by controlling the driving current with a step-shaped signal. The acquisition time for each point is 500 ms, during which an average on both the amplitude and frequency measurements is performed. A typical acquisition contains 100 points, for an overall acquisition time of 50 s. The error in determining the frequency of each point is due to the frequency fluctuations of the free-running QCL during the 500 ms single-point acquisition time, and in our case is ±2 MHz. Reducing this time would imply a better frequency resolution; here we have been limited by the speed of the spectrum analyzer and the data transfer via a GPIB.

Several spectra have been acquired for the same transition, even at slightly different gas pressures. Each set of data has been fitted with a Voigt profile (Fig. 3) to determine the corresponding line-center frequency, whose average value results in $\nu_c = 67678.415$ GHz, with a spread included in the ±2 MHz range. This is in agreement with the experimental error in the frequency measurement that we have mentioned above. The result is consistent with the value provided by the HITRAN database[16] ($\nu = 67678.413$ GHz, with an error between 3 and 30 MHz).

Along with frequency jitter, the QCL temperature fluctuations and the cryostat mechanical instabilities also affect the measurement with amplitude oscillations. These are, at the moment, the main limits to the accuracy of our test. As natural upgrades we are planning both a frequency stabilization, for example, by optically locking the QCL to an external resonance cavity, and a new low-vibration mechanical arrangement.

In conclusion, the first OFCS-referenced QCL to our knowledge has been reported. This result represents a convenient way to extend the OFCS spectral coverage to the MIR; QCLs in fact can ensure low noise combined with high-power outputs, thus allowing high-sensitivity detection levels. Several Doppler-broadened profiles of a $^{13}CO_2$ transition have been acquired, and line-center absolute frequencies measured with a relative precision of 3×10^{-8}.

The reported precision can be heavily improved (at least 3 orders of magnitude) by proper frequency stabilization of the QCL. This upgrade will match the implementation of high-precision spectroscopic techniques such as Doppler-free detection. A big potential of the described apparatus lies in the possibility of directly acquiring the bare QCL emission spectrum, thus making an in-depth analysis of its linewidth possible.

We thank M. Prevedelli (University of Bologna) for developing the phase-lock electronics and L. Nasti for her collaboration on the experiment. This research was partially supported by the RBNE01KZ94 MIUR-FIRB project and by AMRA Scarl.

References

1. T. Udem, J. Reichert, R. Holzwarth, and T. W. Hänsch, Phys. Rev. Lett. **82**, 3568 (1999).
2. S. M. Foreman, D. J. Jones, and J. Ye, Opt. Lett. **28**, 370 (2003).
3. S. M. Foreman, A. Marian, J. Ye, E. A. Petrukhin, M. A. Gubin, O. D. Mücke, F. N. C. Wong, E. P. Ippen, and F. X. Kärtner, Opt. Lett. **30**, 570 (2005).
4. D. Mazzotti, P. Cancio, G. Giusfredi, P. De Natale, and M. Prevedelli, Opt. Lett. **30**, 997 (2005).
5. S. Borri, P. Cancio, P. De Natale, G. Giusfredi, D. Mazzotti, and F. Tamassia, Appl. Phys. B **76**, 473 (2003).
6. P. Maddaloni, P. Malara, G. Gagliardi, and P. De Natale, Appl. Phys. B **85**, 219 (2006).
7. J. Faist, F. Capasso, D. L. Sivco, C. Sirtori, A. L. Hutchinson, and A. Y. Cho, Science **264**, 553 (1994).
8. B. Paldus, C. C. Harb, T. G. Spence, R. N. Zare, C. Gmachl, F. Capasso, D. L. Sivco, J. N. Baillargeon, A. L. Hutchinson, and A. Y. Cho, Opt. Lett. **25**, 666 (2000).
9. G. Gagliardi, F. Tamassia, P. De Natale, C. Gmachl, F. Capasso, D. L. Sivco, J. N. Baillargeon, A. L. Hutchinson, and A. Y. Cho, Eur. Phys. J. D **19**, 327 (2002).
10. S. Borri, S. Bartalini, P. De Natale, M. Inguscio, C. Gmachl, F. Capasso, D. L. Sivco, and A. Y. Cho, Appl. Phys. B **85**, 223 (2006).
11. A. Castrillo, E. De Tommasi, L. Gianfrani, L. Sirigu, and J. Faist, Opt. Lett. **31**, 3040 (2006).
12. R. W. Boyd, *Nonlinear Optics* (Academic, 2003).
13. J. Stenger, H. Schnatz, C. Tamm, and H. R. Telle, Phys. Rev. Lett. **88**, 073601 (2002).
14. T. L. Myers, R. M. Williams, M. S. Taubman, C. Gmachl, F. Capasso, D. L. Sivco, J. N. Baillargeon, and A. Y. Cho, Opt. Lett. **27**, 170 (2002).
15. R. M. 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).
16. HITRAN database, http://cfa-www.harvard.edu/HITRAN.