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Difference-frequency generation in PPLN at 4.25 μm: an analysis of sensitivity limits for DFG spectrometers

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Abstract. We report difference-frequency generation (DFG) in periodically poled lithium niobate (PPLN) around 4.25 μm using a cw Nd:YAG and an injection-locked diode laser. This system provides a narrow linewidth source at 4.25 μm with near-shot-noise-limited operation. A conversion efficiency close to the theoretical limit is obtained. Detection of CO_2 absorption spectra is demonstrated and further improvements and applications to high sensitivity spectroscopy are discussed.

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The availability of quasi-phase-matched (QPM) non-linear crystals, having increased frequency conversion efficiency, has represented a breakthrough for the realization of cw infrared tunable sources. The 3–11 μm region in fundamental rovibrational transitions of most molecules are found and have line strengths at least 1–2 orders of magnitude larger than overtone transitions. If suitable IR sources are available, the IR region can be the best choice to maximize detection sensitivity for molecular detection. The other requirement for high sensitivity is to approach a quantum-noise-limited regime for the spectrometer. QPM crystals can offer an efficiency up to twenty times larger than birefringent devices, just considering the increased effective non-linear coefficient available, and even more if other contributions, such as walk-off angle, are taken into account. Therefore, difference-frequency generation (DFG) can be done in single-pass geometry [1, 2] to produce IR powers in the microwatt range, which is sufficient for high-sensitivity acquisition of molecular spectra.

Room-temperature diode lasers can be used as pump sources to provide wide IR tunability. Extended-cavity diode lasers (ECDLs) are commonly used to get wide reliable tuning. However, ECDL sources have limited output power. DFG spectrometers using ECDLs as the master laser in combination with fiber amplifiers [3] and semiconductor amplifiers [4] have been used for increased output powers. Presently, fiber amplifiers have a wavelength range limited to the 1.5-μm telecommunication band (erbium-ytterbium-doped fibers) or the 0.9–1.1 μm interval (ytterbium-doped fibers) [5].

The alternative that we use in the present system is a pumping scheme based on a cw Nd:YAG laser and a diode laser injection-locked by an ECDL. This allows us to achieve a narrow linewidth and a wide tunability in the IR. We present a careful analysis of the noise for such a source and discuss the sensitivity limits of present-day DFG spectrometers.

1 Experimental setup

Our experimental setup is shown in Fig. 1. Radiation from a diode-pumped, monolithic Nd:YAG laser at 1.064 μm with maximum output power of 800 mW, is coupled onto a dichroic beamsplitter together with the 852-nm radiation from the diode laser system. A 100-mm-focal-length lens focuses the bichromatic beam at normal incidence onto the z-cut lithium niobate crystal ($L = 17.5$, $W = 10$, $H = 0.5$ mm) that was poled at NIST with a domain period of 22.0 μm, using the electric field poling method. Given the domain period, optimum efficiency for 4.25-μm generation was found at a temperature of 287 °C. This PPLN period was originally designed for 3.4-μm generation, but heating allowed us to reach 4.25 μm. However room-temperature mixing for our input wavelengths could be achieved with 23.0-μm poling period. To reach this phase-matching temperature the crystal was contained in a small temperature-controlled copper oven. A PTFE cover ensured good thermal insulation. Radiation at 4.25 μm generated in the crystal is collected with a 50-mm-focal-length CaF_2 lens and focused on the liquid-N_2-cooled InSb detector. The injection-locked diode laser system is composed of a 50-mW master laser mounted in a Littrow extended-cavity configuration [6]; the zeroth-order reflection from the grating injects a 150-mW slave laser. Mode hopping and longitudinal mode instability generally occur for this Fabry–Pérot index-guided laser diode, when unseeded. At maximum emitted power, the slave laser, which is not provided with antireflection coating on the out-

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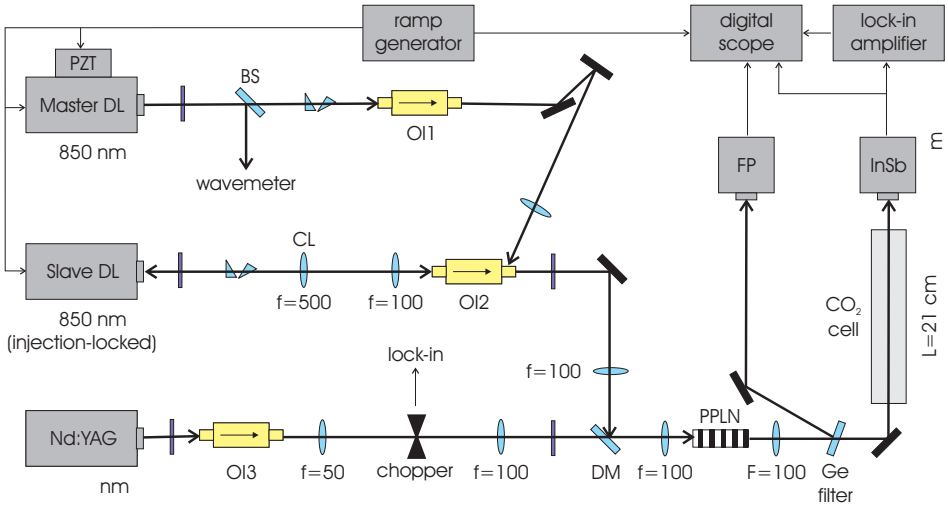

Fig. 1. Experimental setup. $\lambda/2-\lambda/4=$ waveplates; DM = dichroic mirror; FP = Fabry–Pérot cavity; BS = beamsplitter; OI1 = 30-dB optical isolator; OI2–OI3 = 40-dB optical isolators. A $f = 500$-mm cylindrical lens (CL) was used to correct the astigmatism of the slave laser. The master diode laser injects the slave through the isolator OI2, used as optical circulator

put facet, is phase-locked to the master at injected powers larger than 2 mW, with a wavelength-locking bandwidth of about 1.3 nm. We expect that about 99% of the slave laser power will be within the master laser linewidth [7]. This linewidth is a few hundred kHz, as measured in [6], where the same design of extended cavity was characterized. Since the Nd:YAG laser, having a diode-pumped monolithic non-planar miniature ring cavity, contributes a few kHz of linewidth [8], the fast-linewidth (tens of ms timescale) of the IR radiation is determined by the linewidth of the diode laser system. It is worth noting that transitions of the CO_2 band at 4.25 μm have Doppler linewidths of 130 MHz (FWHM) and Lorentzian sub-Doppler width of a few hundred kHz (at pressures of tens of mTorr). This means that precise lineshape broadening studies can be performed with this set- up, as well as saturation spectroscopy if sufficient power is generated. Very recently, sub-Doppler CO_2 transitions, with a linewidth still limited by transit-time broadening have been observed with our setup [9], confirming that the IR power and the linewidth over seconds are sufficient even for the most demanding applications of high-resolution spectroscopy.

2 Noise analysis

We analysed the noise in our system by measuring the amplitude noise of the two pump sources. As expected, at low Fourier-frequency the dominant noise was from the Nd:YAG laser, even if it was equipped with noise-suppressing circuitry designed to reduce the relaxation oscillation peak at about 500 kHz. The measured noise spectral density of the 4.25-μm output beam is shown in Fig. 2. Trace (a) was recorded when the slave diode laser was not injected from the master laser and significantly higher noise can be seen when compared to trace (b), where injection from the master was present. All lasers had the same, fixed, injection current and temperature during the recording of the two traces. It should be remarked that the spectral quality of the pump and signal lasers did not affect the IR generation efficiency and that it is generally not possible to achieve continuous frequency tuning of the unseeded slave laser. Since the IR power was the same in both cases, we attribute the higher noise in trace (a) to mode competition within the non-injected slave laser, which can produce strong amplitude fluctuations. Mode jumps were also evident when analyzing the emission with a 10-MHz linewidth Fabry–Pérot cavity. Due to the intrinsic instability of the slave laser, trace (a) was not reproducible at all, whereas trace (b) was perfectly stable as long as injection was present.

We investigated the predominant amplitude noise sources on our DFG signal. Noise at the output of the detector can be expressed as a function of the current flowing in the detector (which is proportional to the incident power) in the following way [10]:

$$N = P_2 i^2 + P_1 i + P_0 \ , \tag{1}$$

where N is the noise power, i is the current flowing in the detector and the constant term P_0 takes into account detector intrinsic noise and thermal background noise. The linear term results from shot noise and the quadratic term corresponds to the classical intensity fluctuations of the laser. Figure 3 shows experimental measurements of N as a function

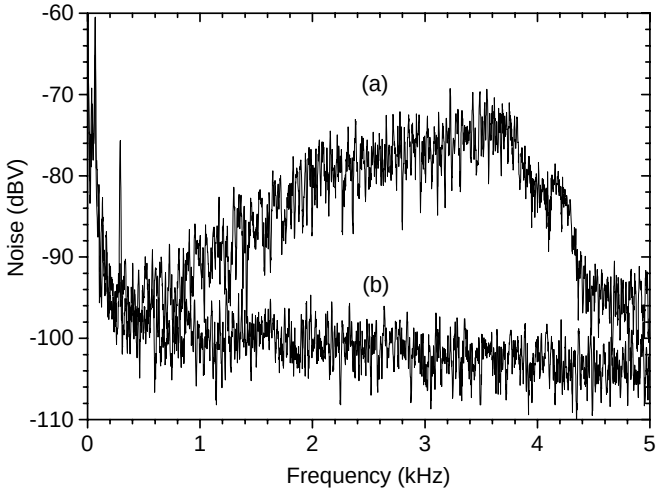

Fig. 2. Amplitude noise measured on the 4.25-μm beam with the InSb detector. Slave laser is not injected in trace (a) and injection-locked in (b). Noise spectra were obtained as FFT of time domain data, acquired by a digital oscilloscope

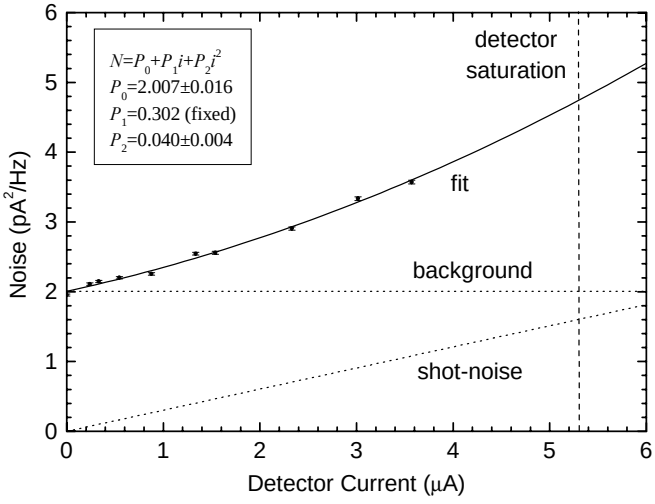

Fig. 3. Experimental points and parabolic best fit curve (see (1)) for amplitude noise density relative to detected photocurrent. Data were taken using a spectrum analyser with 30 Hz resolution bandwidth, 100 s acquisition time, 5 kHz frequency span

of detector current i. Each point is the average of 620 values taken in the 2–5 kHz interval, where the noise is spectrally flat. The small P_2 term, in the fit to the experimental data, confirms that, according to (1), our DFG IR source operates in a near-shot-noise-limited regime. Indeed, at the maximum power allowed by the detector dynamic range, the departure from the shot noise corresponds to 4.7 dB. We attribute the residual quadratic term to Nd:YAG noise transferred to the IR and, partly, to acoustic vibrations acting on the master laser extended cavity. For a better determination of the quadratic term, the P_1 value was fixed, in the fit, at the calculated shot-noise slope. The non-zero intersection with the ordinate axis is then a measure of the P_0 term. Two terms contribute to this constant background: one is the intrinsic detector-noise and the other is the thermal fluctuation of the background radiation reaching the detector. By putting a reflecting mirror in front of the detector, so that the liquid-N_2-cooled detector looks at a background that is close to its own temperature (77 K), we verified that 60% of the noise at zero current comes from thermal fluctuations. A similar measurement, without the reflecting mirror, but with the oven at the working temperature, showed that only 9% additional noise comes from the hot oven, due to the small solid angle seen by the detector (about 10^{-3} sr). It would be possible to reduce the departure from the shot noise to 2.9 dB, if thermal fluctuations were efficiently reduced by use of proper narrow-bandwidth cold filters.

3 Spectroscopic performance

The spectrometer performance was tested by recording several rovibrational lines of $^{12}C^{16}O_2$ belonging to the ν_3 band. In Fig. 4, the R(82) line is shown, recorded, in a single scan, by filling a 21-cm-long cell, equipped with CaF_2 windows, to a pressure of 1.33 kPa pure CO_2. The horizontal axis in Fig. 4 shows a 0.9-GHz scan of the DFG frequency. This was obtained by feeding a 3.2-V_{p-p} ramp to the piezoelectric ceramic (PZT) on the feedback grating of the master laser. In order to avoid mode jumps, a proportional voltage was synchronously fed to the modulation input of the master injection-current as well as to the slave laser current. In this configuration we achieved a span of 12 GHz, limited only by the maximum output voltage of the ramp generator. The experimental data shown in this figure are fit with a Voigt profile for the stronger line and a sinusoidal modulation of the baseline, due to optical fringes in our apparatus. The Lorentzian profile due to the small CO_2 atmospheric absorption is much larger (about 3.5 GHz FWHM) than the frequency span and its contribution, as well as laser power modulation due to the voltage ramp, is taken into account in the baseline fitting with linear and quadratic terms. As is shown in Fig. 1, the Nd:YAG beam was intensity modulated by a chopper operating around 1 kHz. The InSb detector, followed by an analog lock-in amplifier, allowed phase-sensitive detection of the modulated 4.25-μm beam. From the fit of the experimental data to a Voigt profile with a fixed Doppler contribution of 66.2 MHz (HWHM), we obtain a Lorentzian contribution of 21.5 MHz (HWHM). This Lorentzian width is in good agreement with the value (about 23 MHz) that can be inferred from the HITRAN database [11]. This database predicts a peak absorption, expressed as αL (α = peak absorption coefficient; L = cell length), that is about 12% larger than our experimental value. Detector calibration performed with a black-body source, using piecewise integration of the detector responsivity against the black-body curve, gives a responsivity of $R = 2.7$ A/W at 4.25 μm. To record and identify absorption lines, wavelength measurements were performed using a wavemeter with a resolution of a few parts in 10^7 on the diode laser beam.

After correction of the astigmatism of the slave laser (with a cylindrical lens), the DFG power was increased by about a factor of 2.5, giving a maximum IR power of 12 μW, with 80 and 710 mW incident on the crystal from the slave laser and the Nd:YAG laser, respectively. This result translates into an efficiency of 0.021%/W, which compares well with the previous results (0.010%/W) at this wavelength [1]. The theoretical limit is 24 μW (0.042%/W efficiency), calculated from the relation given in [1], with $d_{\text{eff}} = 14$ pm/V and taking into account the Fresnel reflection losses from

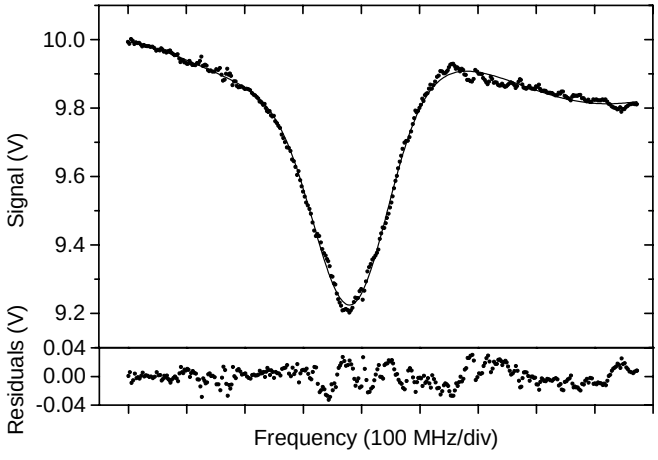

Fig. 4. Experimental recording of R(82) $^{12}C^{16}O_2$ line. The cell length was 21 cm, the CO_2 pressure 1.33 kPa, the detected photocurrent 0.5 μA, the lock-in time constant 10 ms, and the scan time 20 s. The *fit curve* is superimposed on data points and the plot of residuals is shown below it

crystal surfaces, 13.8%, 13.4%, and 11.8%, respectively, for 850 nm, 1064 nm, and 4.25 μm beams. We attribute the discrepancy between experimental and calculated efficiency to a non-optimized focusing for input beams. Moreover, it is likely that the duty cycle of the poling period is not precisely 50%, which will also reduce the efficiency of the non-linear mixing.

4 Summary

From the recording reported in Fig. 4, a $\Delta I/I$ of about 10^{-3} can be calculated. This number is in striking contrast with the limiting sensitivity $\Delta I/I$ of 2.5×10^{-7} (1 Hz bandwidth and $R = 2.7$ A/W) that could be obtained, under shot-noise-limited conditions, using the liquid-N_2-cooled InSb detector mounted in our setup, saturating at 1.9 μW incident power. From data in Fig. 3, it comes out that our DFG source is 4.7 dB above shot-noise (corresponding to a sensitivity of 4×10^{-7}). Whereas in this figure each point is the average of noise values in the 2–5 kHz range, the absorption line reported in Fig. 4 has predominant noise contributions from very low Fourier frequencies. Therefore, the signal-to-noise ratio is significantly degraded, for the CO_2 recording, by excess technical noise, mainly due to optical interference fringes and to thermal and mechanical instabilities. In order to fully exploit the spectrometer sensitivity, scan times that are in the ms range should be selected, to efficiently reject Fourier frequencies lower than kHz. In our present setup, we are limited by the maximum chopping frequency (4 kHz) and by the lock-in minimum integration time (1 ms). It should be noted that, even if baseline modulation is generally introduced by wavelength scanning, sub-shot-noise spectroscopy was demonstrated [12] using the same master-slave diode laser system we use in the present work.

In conclusion, we have demonstrated operation of a new DFG spectrometer, which may become a powerful tool for high-resolution spectroscopy. In particular, the reasonable IR level power, combined with the very narrow linewidth and low amplitude noise performance, is well suited for sub-Doppler spectroscopy of molecular transitions. Frequency locking onto narrow resonances in the infrared could also be useful to bridge the gap between near- and mid-IR regions for metrological purposes.

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