5 μm laser source for frequency metrology based on difference frequency generation

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A narrow-linewidth cw 5 μm source based on difference frequency generation of a 1.3 μm quantum dot external cavity diode laser and a cw Nd:YAG laser in periodically poled MgO : $\mathrm{LiNbO_3}$ has been developed and evaluated for spectroscopic applications. The source can be tuned to any frequency in the 5.09–5.13 μm range with an output power up to 0.1 mW, and in the 5.42–5.48 μm range with sub-microwatt output. The output frequency is stabilized and its value determined by measuring the frequency of the two lasers with a remotely located frequency comb. A frequency instability of less than 4 kHz for long integration times and a linewidth smaller than 700 kHz were obtained. © 2012 Optical Society of America

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The mid-IR spectral range offers a plethora of molecular vibrational transitions with very small transition linewidths, due to the long spontaneous lifetimes. These transitions are especially interesting when the molecules are cold, as is achievable with several methods [1]. While direct spectroscopy with a mid-IR comb [2,3] might be possible, often it will be more practical or necessary to use a sufficiently powerful single-frequency source for molecule excitation. It should be frequency-stable, exhibit narrow linewidth, and its frequency should be measurable with respect to a primary frequency standard.

In one approach, a quantum cascade laser is the spectroscopy source, and its radiation is upconverted into the near-IR for stabilization to and measurement by a conventional fiber frequency comb [4,5]. In Ref. [5], a quantum cascade laser (4.3 μm) was stabilized to a linewidth of 6.8 MHz. Another approach, capable of satisfying the requirements above, employs a parametric downconversion process of one or two lasers with relatively short wavelength. Suitable lasers with narrow free-running linewidth are available (e.g., external cavity diode lasers, solid-state lasers, fiber lasers), and their spectral range overlaps with current femtosecond frequency combs of the Ti:sapphire or Er:fiber type. The parametric downconversion can be a difference frequency generation (DFG) of two lasers or an optical parametric oscillation (OPO). The two implementations both have their advantages and disadvantages. In both cases, the (simultaneous) measurement of the frequencies of two waves must be carried out, that is, of the two input laser frequencies (DFG), or of the pump laser and signal wave frequencies (OPO), respectively. Examples of these approaches are given in Refs. [6–10], where the longest wavelength reached has been 4.57 μm [9]. This is due to the limiting effect of the intrinsic absorption of the typically used oxide crystals beyond 4.5 μm [10].

In this work, we have extended the upper wavelength limit of difference frequency generation into the strongly absorbing spectral range. By resonant enhancement of one of the two lasers we have nevertheless achieved a reasonable output power level for spectroscopy, up to 0.1 mW, even at 5.1 μm. One employed laser is a novel quantum dot external cavity diode laser (QD-ECDL) [11], the other is a monolithic cw Nd:YAG laser, frequency-stabilized to a hyperfine transition in molecular iodine. The DFG output wavelength is tunable from 5.09 to 5.13 μm by simultaneous tuning of the diode laser wavelength and of the MgO:PPLN crystal temperature. Absolute frequency measurement of the mid-IR radiation is done indirectly by a hydrogen-maser/global positioning system (GPS)-referenced Ti:sapphire frequency comb that measures simultaneously the 1.06 and 1.34 μm wave frequencies.

Figure 1 shows the experimental setup. A high power (12 W, 1064 nm) cw Nd:YAG master-oscillator/slave laser (Innolight/Laser Zentrum Hannover) is used as pump source. The laser is stabilized to a rovibrational transition of molecular iodine via Doppler-free saturation spectroscopy. The maximum power available in front of the nonlinear crystal is 5.7 W. The signal wave source is a home-built QD-ECDL (Innolume LD:GC-1320-CM-200) in Littrow configuration with a free-running linewidth of 5 MHz. The laser is tunable in the range of 1316–1345 nm with an output power of 45 mW at 1344 nm, leading to a usable power after the optical isolators (one bulk and one fiber-based, totaling –90 dB isolation) of about

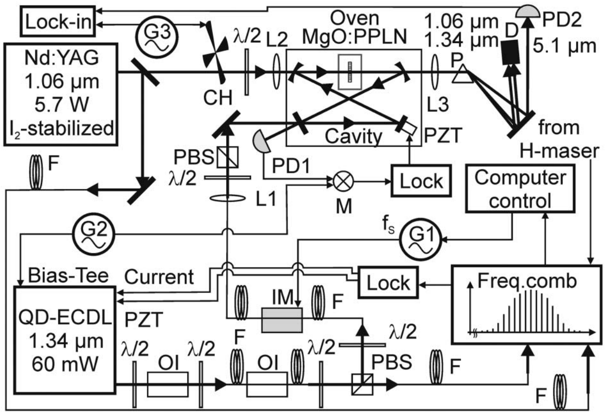

Fig. 1. Experimental setup: $\lambda/2$ retardation plates; OI, optical isolators; PBS, polarizing beam splitter cubes; F, single-mode optical fibers; IM, intensity modulator; CH, chopper wheel (for lock-in detection); PD, photodetector; P, prism; L, lenses; G, r.f. oscillator; PZT, mirror actuator; D, beam dump; M, mixer.

19 mW. To enhance the power of this laser source, a cavity, singly resonant for 1344 nm, is used. The cavity length is stabilized to the diode laser's frequency using the Pound-Drever-Hall method. With a finesse of 200, an enhancement factor of up to 60 was achieved, leading to a circulating power of 0.9 W at 1344 nm.

The MgO:PPLN crystal (HCP Photonics) is 7.4 mm long, 0.5 mm thick, 10 mm wide, and possesses six different poling periods in the range of 21–25.4 μm. Crystal faces are polished plane-parallel and antireflection (AR)-coated. The crystal's temperature is adjustable between 20–100°C, using a Peltier element driven oven. The attenuation coefficient α of the MgO:PPLN was independently measured to be $\alpha = 6.75\ \mathrm{cm}^{-1}$ at 5.485 μm and is a significant limiting factor on the achievable output power in the spectral range around 5 μm. Pump and signal beams are focused by lenses L1 (fiber collimator) and L2 (175 mm focal length) into the $\Lambda = 25.4\ \mu$m poling period section of the crystal. In order to minimize the absorption of the generated 5 μm radiation, the foci of the two lasers, and therefore the region of largest DFG power generation, are close to the exit face of the crystal. The working point for best phase matching is 71°C, resulting in a DFG output power up to 105 μW at 5.115 μm. The mid-IR output is collimated and focused using an AR-coated $\mathrm{CaF_2}$ lens (L3). The pump and signal beams are separated from the idler beam using a $\mathrm{CaF_2}$ prism (P), which efficiently transmits the p-polarized idler wave. For detection, the 1064 nm wave is chopped and the modulated 5.1 μm wave is sent on a pyroelectric detector (PYRO-LME 353) followed by lock-in detection.

In addition, the poling period $\Lambda = 24.2\ \mu$m was tested for DFG with the diode laser tuned to 1320 nm, and both lasers in single-pass configuration (no cavity), obtaining tunable radiation in the range of 5.42–5.48 μm with 0.1 μW output level.

The frequency of the 1344 nm diode laser is stabilized to and measured by a frequency comb that is also used for frequency measurement of the iodine-stabilized 1064 nm Nd:YAG laser. The frequency comb is based on a Ti:Saphire laser and a Menlo Systems comb kit (FC 8004), modified in-house. The 1344 and 1064 nm waves are sent via two 70 m fibers to the frequency comb lab. Here, after coupling the waves out of the fibers, they are sent into two beat lines, in which the cw waves are overlapped with the comb radiation and their beats detected by fast photodetectors. Appropriate bandpass filters block the unnecessary comb modes before the overlap. The beat between the laser waves and the nearest comb modes are filtered and amplified with tracking oscillators. The frequency comb's repetition rate f_{rep} and carrier envelope frequency f_{CEO} are locked to a hydrogen maser controlled synthesizer, and the maser is itself steered to GPS on long time scales. Figure 2 shows the beat note of the 1344 nm diode laser which is frequency-stabilized to a fixed radio frequency (r.f.) with the aid of a frequency-phase detector [12]. The linewidth of the beat note is $\Gamma_{1344\text{–comb}} \approx 720$ kHz. The linewidth of the iodine-stabilized Nd:YAG laser is $\Gamma_{1064} \approx 150$ kHz, determined by beating with an independent narrow-linewidth Nd:YAG laser. The linewidth of the frequency comb modes is $\Gamma_{\mathrm{comb}} \approx 272$ kHz, also determined from a beat with the independent Nd:YAG laser. Therefore, the linewidth of the 5.1 μm radiation is approximately

$$\Gamma_f = (\Gamma^2_{1064} + \Gamma^2_{1344\text{–comb}} - \Gamma^2_{\mathrm{comb}})^{1/2} \approx 0.68\ \mathrm{MHz}.$$

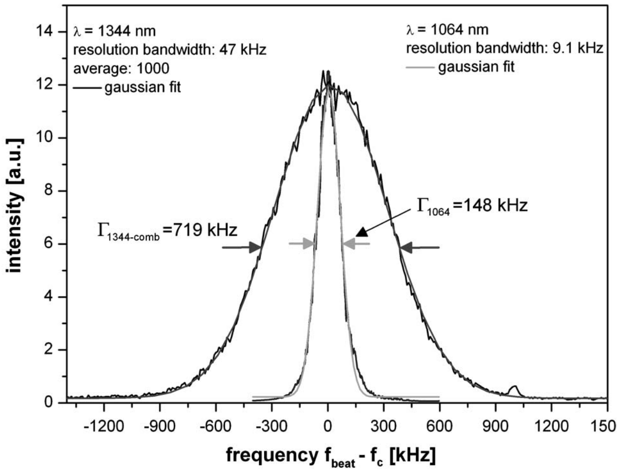

Fig. 2. Beat notes between the 1344 nm diode laser and the frequency comb and between the 1064 nm Nd:YAG laser and a narrow-linewidth (10 kHz) Nd:YAG laser (ILF100, Institute for Laser Physics, Novosibirsk).

The beat notes are counted by a dead-time-free frequency counter and logged together with the frequency comb's repetition rate f_{rep} and the calculated DFG frequency. Figure 3 shows the time traces of the Nd:YAG and the diode laser frequencies, measured with the frequency comb and the calculated difference frequency at 5.1 μm. Its frequency instability is mostly due to that of the Nd:YAG laser, because of limitations of the iodine stabilization. The Allan deviation is shown in Fig. 4. The frequency instability is less than 25 kHz for short integration times and drops to 4 kHz for an integration time above 200 s.

Tuning of the generated difference frequency at 5.1 μm can be done by variation of the frequency comb's repetition rate f_{rep}, while all systems remain in lock. Typically,

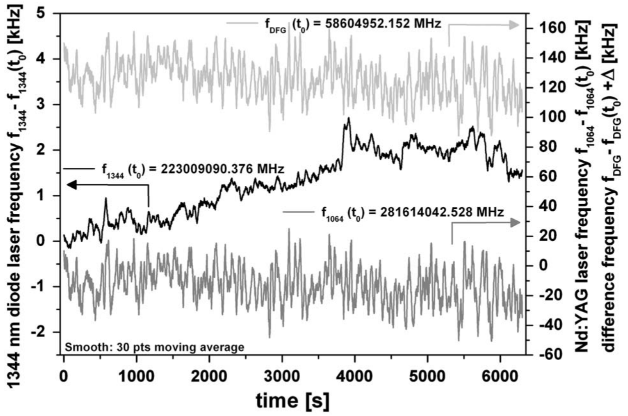

Fig. 3. Frequency traces of the Nd:YAG laser and of the diode laser, measured by the frequency comb over more than 1 h and the corresponding frequency trace of the generated difference frequency at 5.1 μm. The DFG frequency trace has a frequency offset of $\Delta = +140$ kHz for illustration purpose. Note the different vertical scales.

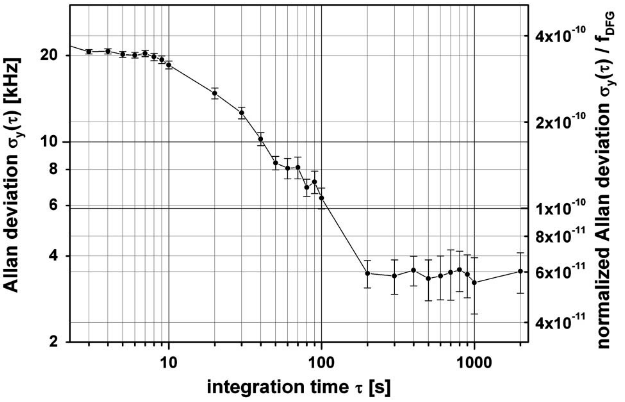

Fig. 4. Allan deviation of the difference frequency values at $5.1\,\mu$m.

a frequency sweep of (only) 10 MHz/ min, depending on the comb's working point, could be achieved.

A second approach for tuning the QD-ECDL's frequency while in lock, and thus the frequency of the $5.1\,\mu$m radiation, has been implemented using a waveguide intensity modulator (Photline Technologies, MX1300-LN-10). The LiNbO$_3$-based, low-insertion-loss intensity modulator is driven by a signal generator at a r.f. f_s and generates two symmetric sidebands ($f_{1344} \pm f_s$) with respect to the carrier frequency f_{1344}, which is kept fixed (stabilized to the comb). For sufficiently high r.f. drive strength, the carrier is suppressed and its power (nearly) completely transferred to the sidebands. The enhancement cavity is locked onto one sideband, e.g., $f_{1344} - f_s$, via the Pound-Drever-Hall technique. The generated $5.1\,\mu$m radiation f_{1064} – ($f_{1344} - f_s$) then has a frequency offset $+f_s$ with respect to the frequency f_{1064}–f_{1344} measured by the comb. The r.f. f_s can be swept in time and the cavity lock system follows the sweep, yielding a frequency sweep in the generated $5.1\,\mu$m radiation. Figure 5 shows a typical frequency sweep. The maximum sweep range for f_s is 15 to 475 MHz, with an output power of about $2.2\,\mu$W at $5.1\,\mu$m. It is possible to manually change to a lock on the opposite sideband, thereby doubling the sweep range. To determine whether the cavity is locked to the positive or negative sideband (and therefore knowing the actual frequency at $5.1\,\mu$m), the voltage applied to the cavity mirror piezo actuator is constantly monitored. When the lock is to the negative sideband "$-f_s$", the corresponding piezo voltage decreases upon increasing the modulation frequency, while the opposite is the case for a lock to the positive sideband "$+f_s$". The maximum frequency tuning rate was 35 MHz/s.

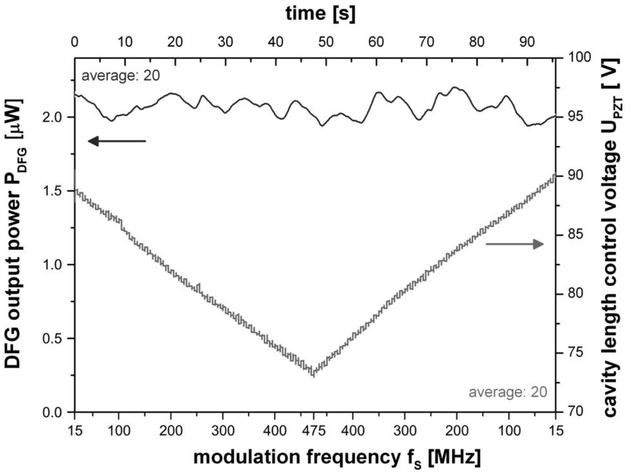

Fig. 5. DFG output power and the cavity length control voltage during a typical frequency sweep using the intensity modulator tuned to the negative sideband "$-f_s$".

This $5.1\,\mu$m source was successfully used in an experiment that observed and measured the frequency of the most fundamental electric dipole-allowed molecular vibrational transition, the $v = 0 \rightarrow v = 1$ transition in the molecule HD$^+$ (observed linewidth: 3 MHz) [13].

In conclusion, we have developed a cw narrowband mid-IR source with $105\,\mu$W maximum output power based on DFG between a Nd:YAG laser and a $1.3\,\mu$m QD-ECDL. The source was tunable to any frequency in the 5.09–$5.13\,\mu$m range and is relatively rapidly tunable over a range of 460 MHz, using a sideband generator, with an output power of $2.2\,\mu$W. Stabilized in part and measured by a hydrogen-maser/GPS-referenced Ti:sapphire frequency comb, the generated $5.1\,\mu$m radiation possessed a frequency instability of 8 kHz or less for integration times larger than 10 min and a spectral linewidth smaller than 700 kHz, making it well suitable for precision spectroscopic applications.

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