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Published in:
Optics Letters

DOI:
10.1364/OL.40.002692

Publication date:
2015

Document Version
Peer reviewed version

Link to publication in Heriot-Watt University Research Gateway

Citation for published version (APA):
Mode-resolved 10-GHz frequency comb from a femtosecond optical parametric oscillator

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Received Month X, XXXX; revised Month X, XXXX; accepted Month X, XXXX; posted Month X, XXXX (Doc. ID XXXXX); published Month X, XXXX

We report a 10-GHz frequency comb generated by filtering a 333.3-MHz OPO frequency comb with a Fabry-Pérot (FP) cavity, which was directly stabilized to the incident fundamental comb. This result is supported by a detailed analysis of the Vernier-effect-induced multiple peaks in the transmitted comb power as the FP cavity spacing is detuned. Modes of the generated 10-GHz comb were clearly resolved by a Fourier transform spectrometer with a spectral resolution of 830 MHz, considerably better than the Nyquist sampling limit. The potentially broad tuning range of this mode-resolved OPO frequency comb opens unique opportunities for precise frequency metrology and high-precision spectroscopy.

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http://dx.doi.org/10.1364/OL.99.099999

Since their first demonstration in 2007 [1], frequency combs based on femtosecond optical parametric oscillators (OPOs) have been shown to be versatile devices capable of operation at high average powers [2] and with tunability into the visible [3] and mid-infrared [2]. Notably, they are able to achieve comb linewidths of a few Hz [4] and timing jitters across the comb as low as 30-attoseconds [5]. Their direct extension to multi-GHz repetition rates is however limited by considerations such as the high oscillation threshold associated with low peak power multi-GHz pump lasers and the limited availability of such sources, although harmonic GHz operation of a fully stabilized OPO comb was recently reported [6]. In this Letter we describe the extension of a fully stabilized OPO frequency comb to > 10 GHz mode spacing by use of a Fabry-Pérot (FP) filter cavity directly locked to a subset of the comb modes. We demonstrate the practicality of this approach by clearly resolving around 800 individual filtered comb modes with a resolution many times above the Nyquist sampling limit, exceeding the best previously reported performance in which a spectrometer with a resolution of 210 MHz was used to resolve the modes of an OPO comb with a mode spacing of 417 MHz [7].

The extension of stabilized OPO combs to >10-GHz mode spacings is motivated by both spectrometry and spectroscopy. For example, the calibration of high-resolution echelle spectrographs in astronomy requires laser frequency combs in the 10–30-GHz range [8, 9], and led to the development of several Fabry-Pérot (FP) filtered laser combs and related techniques [10–13]. Future instruments such as the proposed high-resolution spectrograph (HIRES) on the European Extremely Large Telescope (E-ELT) will require calibration sources spanning the 0.37–2.4 µm wavelength range [14], exceeding the range of typical laser combs. At 10-GHz mode spacings and above, it also becomes practical to use grating spectrometers to resolve individual comb lines, which opens the possibility of high resolution direct comb spectroscopy in the mid-infrared, to measure not only the line-centres of absorption features but also their line-shapes, which contain important physics of relevance to modelling the rich absorption spectra of greenhouse gases such as CO₂ and water vapor [15]. Recently, mid-infrared frequency combs have been used in conjunction with tunable single-frequency narrow-linewidth lasers to implement comb-assisted spectroscopy [16–19]. Utilizing the idler output of a multi-GHz OPO frequency comb would allow such precision spectroscopy to be conducted by simply sweeping the repetition frequency of the OPO comb, removing the need for a tunable single-frequency laser.

Our experimental scheme is shown in Fig. 1. The OPO, pumped by a mode-locked Ti: sapphire laser (Laser Quantum Gigajet), was based on a previously reported design [6]. The generated signal comb had a mode-spacing of 333.3 MHz, a spectral coverage over 1.1–1.6 µm by cavity-length or grating tuning, and a pulse width of 60–90 fs across the tuning range. The pump repetition rate was sampled with a photodiode and its 6th harmonic (2GHz) was locked to a 2 GHz reference, which was then referenced to a 10-MHz Rb clock [6]. The carrier-envelope-offset (CEO) frequency of the signal output from the OPO was detected by heterodyning light from the pump supercontinuum with non-phase-matched pump-signal sum-frequency light from the OPO, and also locked to a 10-MHz reference. Details of the locking scheme were reported in [4, 6]. For a one-second gate time, the measured instability in fCEO and frep were 0.27 Hz and 1.5 mHz respectively, and the resultant fractional instability in the comb frequencies was 5 × 10⁻¹².

The FP cavity was composed of two low-dispersion plane mirrors, with 99% reflectivity from 1.35–1.60 µm. The mirrors were also coated with a reflectivity of 99% at 780 nm. One mirror was mounted on a translation stage,
which was used to coarsely adjust the free spectral range (FSR) of the FP cavity. The other mirror was mounted on a piezoelectric transducer (PZT), which was used to dither lock the cavity by modulating it at a frequency of 9.2 kHz and with an amplitude of 0.8 nm (see Fig. 1). A single-frequency external cavity diode laser (ECDL) with a central wavelength at $\sim$780 nm was combined with the OPO beam using a dichroic mirror and simultaneously coupled into the FP cavity to assist the initial angular alignment of the mirrors.

![Fig. 1. Schematic of the experimental setup, including the pump and OPO, a nonlinear interferometer for detecting the CEO frequency ($f_{CEO}$) of the signal or idler, a loop for stabilizing the $f_{CEO}$, a stabilized comb filter, and an FTIR spectrometer. PCF, photonic crystal fiber; pSC, pump supercontinuum; PBS, polarization beam-splitter; IF, interference filter; P, polarizer; APD, avalanche photodiode; PZT, piezoelectric transducer; PI controller, proportional-integral controller; PFD, phase-frequency detector; LPF, low-pass filter; HPF, high-pass filter; Amp, amplifier; PD, photodiode; ECDL, external cavity diode laser.](image)

During alignment, a triangular voltage was applied to the PZT to scan the cavity. The voltage applied to the PZT, and the transmitted power in the ECDL and OPO channels are shown in Fig. 2(a–c). The mirror displacement corresponding to the two transmission peaks (Fig. 2(b)) was 390 nm, half the wavelength of the ECDL. When the cavity length was tuned over this range, 16 transmission peaks were observed in the OPO channel, as expected since the OPO comb is composed of many evenly-spaced, single-frequency laser lines. When the free spectral range of the FP cavity deviates slightly from the harmonics of the fundamental comb mode-spacing, the overall filtered output power oscillates due to the Vernier effect. To understand this, a simulation was constructed and the result is shown in Fig. 2(d), in which the mode-spacing of the incident comb was set at 333.3 MHz with a central wavelength at 1500 nm, while the FP cavity length was set at ~1.5 cm to support an FSR of 10 GHz (30th harmonic of 333.3 MHz). The simulation shows close agreement with the experimental result shown in Fig. 2(e). The discrepancy in the fringe width, and the difference in the rate of change in fringe intensity between experiment and simulation, we believe are due to imperfect mode-matching of the OPO light into the FP cavity.

Firstly, the OPO signal comb was fully locked, with a mode spacing of 333 MHz and CEO frequency of 10 MHz, and passed through the FP cavity. Then, the FP cavity length was adjusted manually to approximately 1.4 cm, and the transmitted comb power was monitored when a sweep signal was applied [Fig. 2(c)]. A dither locking scheme [12, 20] was then implemented to stabilize the FP cavity to the maximum transmission of the frequency comb, which corresponds to the peak of the trace in Fig. 2(c). In acquiring the RF spectrum shown in Fig. 3, the FP length was approximately 1.4 cm, corresponding to a FSR of 10.67 GHz. The filtered comb had a mode spacing of 10.67 GHz, corresponding to the 32nd order harmonic of the fundamental comb, and the side mode suppression ratio (SMSR) was ~20 dB. Higher SMSR could be obtained by cascading two or more FP cavities [11]. Careful thermal and mechanical isolation provided stable locking, which was even tolerant to the slow drift of the repetition rate of the fundamental comb when this was free running.

The powers of the fundamental and harmonic combs were measured to be 20 mW and 0.3 mW respectively, implying a FP transmission of 1.5%. In addition to the 96.9% loss due to the spectral filtering from 333 MHz to 10.67 GHz, the remaining loss is attributed to the...
imperfect mode-matching, since plane mirrors were used for building the cavity. Spectral narrowing can be ruled out since the OPO spectra measured before and after the FP cavity were nearly identical.

The 10-GHz OPO frequency comb was characterized by using a high-resolution mode-resolving Fourier-transform spectrometer, constructed according to the design shown in Fig. 1. It utilized two retroreflectors mounted on a long-travel motorized stage, which dynamically changed the optical path difference (OPD) of the two arms of the interferometer simultaneously and in opposite directions. The travel range of the motorized stage used in the spectrometer was 10 cm, giving an OPD of 40 cm, which supports a spectral resolution of 0.025 cm⁻¹ (750 MHz) [21]. A narrow-linewidth HeNe laser with a central wavelength at 632.816 nm was also coupled into the interferometer for absolute delay calibration. A silicon photodiode (omitted for clarity in Fig. 1) was used to detect the HeNe output from the interferometer, while an InGaAs photo-detector was used to measure the comb. Both channels were sampled by a 16-bit digital acquisition card at a sampling rate of 2 megasamples per second.

The generated interferogram recorded by scanning the Fourier-transform spectrometer stage over 10 cm is shown in Fig. 4. The measurement time was 1.5 seconds and the OPD was calibrated using a simultaneously recorded HeNe laser interferogram. For the purposes of calculating the comb spectrum, the interferogram was windowed to an optical path length of 36 cm, which supports a spectral resolution of 0.83 GHz (0.028 cm⁻¹). In these results, recorded with the FP cavity length set for 31st order filtering, the mode-spacing of the filtered comb was 10.33 GHz, corresponding to a temporal pulse spacing of 97 ps. For this reason, the recorded interferogram (Fig. 4(a)) contains 13 interference maxima across its 1200-ps scan range, one of which is shown in Fig. 4(b-c) on a zoomed scale. Due to weak reflections from the low-reflectivity surface of the beam splitter used in the spectrometer, there are satellites near all the main interference maxima; we believe these artifacts can be removed by using a wedged beam splitter. For clarity, we manually removed these satellites before the spectrum was computed.
Figure 5(a) shows the resulting spectrum spanning >8 THz, which was obtained by Fourier transforming the interferogram in Fig. 4(a). The 800 individual comb lines are resolved across the full spectral span, as shown in the zoomed spectra in Fig. 5(b, c), and the mode spacing of 10.3 GHz is clearly seen in Fig. 5(d). The apodized instrumental linewidth was 1.0 GHz. The signal-to-noise ratio (SNR), defined as the ratio between the peak power of an individual comb line and the average noise power between two adjacent comb lines over 1 GHz, was 13 dB near the centre wavelength of the comb source.

This demonstration of mode-resolved FP filtering of an OPO frequency comb employed the near-infrared signal output from the OPO, but equivalent performance can be expected in the mid-infrared by using the idler comb, representing an ideal source for molecular spectroscopy [22]. Indeed, the idler output from this OPO can be tuned to cover 2-4 μm mid-infrared region, which is important for sensing molecules. Femtosecond OPO based FTIR spectroscopy has already been demonstrated with high sensitivity, high acquisition rate, high resolution, and the capability of standoff sensing [23–25], and can be extended to exceptionally high resolution by using dual combs with slightly different mode spacings [26–28]. The ability to spectrally resolve individual OPO comb modes now allows the extremely narrow Hz-level comb linewidth [4] to be fully exploited to conduct precision spectroscopy of narrow molecular absorption features over the full OPO wavelength tuning range.

In summary, we have demonstrated a laser frequency comb with a mode spacing of >10 GHz by filtering an OPO comb with a Fabry-Pérot cavity, which was directly stabilized to the incident fundamental comb. Individual modes of the filtered comb were clearly resolved by using a Fourier-transform spectrometer with a resolution of 0.83 GHz. The comb-mode-resolved frequency comb from a Fourier-transform spectrometer with a resolution of 10.3 GHz is clearly seen in Fig. 5(d). The apodized zoomed spectra in Fig. 5(b, c), and the mode spacing of 10.3 GHz is clearly seen in Fig. 5(d).

This research was carried out under the METROCOMB project, which has received funding from the European Community’s Seventh Framework Programme (FP7/2007-2013) under Grant Agreement no. 605057. The authors are also grateful for funding from the Science and Technology Facilities Council (STFC ST/L002140/1).

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