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# High-power asynchronous midinfrared optical parametric oscillator frequency combs

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We introduce a new stabilization scheme providing a pair of high-power, carrier-envelope-offset (CEO) frequency-stabilized, broadband, asynchronous frequency combs operating at 3.3  $\mu\text{m}$ . The two channels, each with 100 mW average power and originating from a single synchronously pumped optical parametric oscillator, share all the components for midinfrared generation and CEO-frequency detection, and can be stabilized independently at repetition frequencies up to 5 kHz apart. This unique source is fully compatible with midinfrared dual-comb spectroscopy, and the approach can be readily extended to other wavelengths. © 2013 Optical Society of America

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In a dual-comb spectrometer, two asynchronous laser frequency combs with different pulse repetition rates ( $f_{\text{rep}}$ ) and stabilized carrier-envelope-offset (CEO) frequencies ( $f_{\text{CEO}}$ ) are mixed before or after a sample, yielding a down-converted radio-frequency (RF) comb that contains information on the absorption experienced by the combs [1–5]. The absence of moving parts in such a spectrometer overcomes the speed and resolution limitations of a scanning Michelson interferometer. Its resolution is only limited by the comb-line spacings, and the recording time is the inverse of the difference in the pulse repetition rates of combs. To date, most dual-comb spectroscopy has been conducted in the near-infrared [1,2], but for trace-gas detection and analysis it is essential to extend coverage to the midinfrared (mid-IR) region, which is the fundamental absorption band for many molecules.

Mid-IR frequency combs can be generated by either difference frequency generation (DFG) schemes or optical parametric oscillators (OPOs). Until now, all existing asynchronous mid-IR (2.5–20  $\mu\text{m}$ ) frequency combs have been based on low-efficiency DFG processes, and the generated powers were no more than 1 mW [3,4], significantly limiting the signal-to-noise ratio in the mid-IR measurement. By contrast, a synchronously pumped OPO (SPOPO) has far superior conversion efficiency, and recently we introduced a method for generating two separate mid-IR pulse sequences with nearly identical spectral properties and slightly different repetition rates from a single SPOPO, pumped by two independent lasers [6]. The two OPO oscillation channels, with a stabilized repetition-rate difference of up to 5 kHz, generated broadband mid-IR (3.3  $\mu\text{m}$ ) idler spectra with an average power of 100 mW from each channel. In this Letter, we report on the detection and stabilization of the CEO frequencies of the two asynchronous channels simultaneously. This represents a major step forward, providing for the first time a pair of practical and high-power phase-coherent asynchronous mid-IR frequency combs, with sufficient powers for dual-comb stand-off spectroscopy.

The layout of the system for generating and stabilizing the asynchronous mid-IR combs is shown in Fig. 1. The pump sources and OPO were based on the designs in [6,7]. The two pumps, each composed of a fiber amplifier seeded by a mode-locked Yb:KYW laser, were identical except for their repetition rates, and were combined on a 50:50 dielectric beam-splitter. The average power, center wavelength, –3 dB spectral bandwidth, and pulse-width of each pump source were 2.5 W, 1058 nm, 10 nm (4.82 THz), and 3 ps, respectively. The output from one port of the 50:50 beam-splitter was used to pump the SPOPO, generating two idler channels of broadband

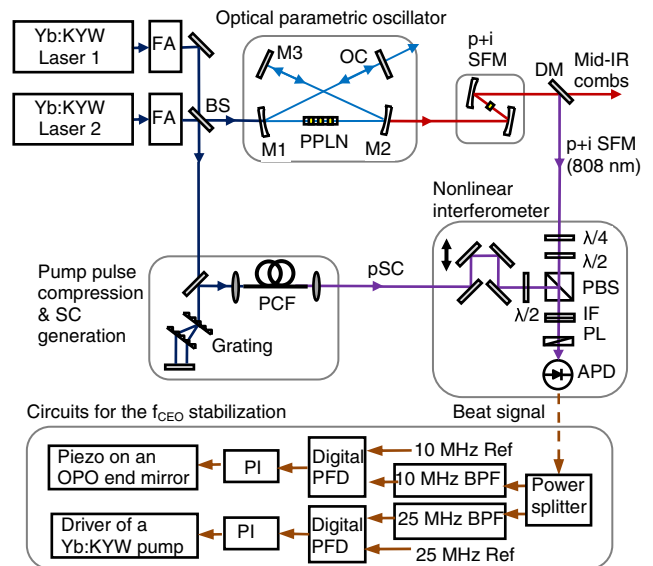


Fig. 1. Layout of the asynchronous mid-IR combs. FA, fiber amplifier; BS, beam-splitter; M, mirror; OC, output coupler; SFM, sum-frequency mixing; DM, dichroic mirror; PCF, photonic crystal fiber; pSC, pump supercontinuum; PBS, polarizing beam-splitter; IF, interference filter; PL, polarizer; APD, avalanche photodiode; BPF, band-pass filter; PFD, phase-frequency detector; PI, proportional-integral amplifier.

mid-IR spectra (3.3  $\mu\text{m}$ ,  $-3$  dB bandwidth  $>170$  nm), each with an average power of  $\sim 100$  mW, and with a stabilized repetition-rate difference of up to 5 kHz.

The idler  $f_{\text{CEO}}$  was detected by heterodyning the pump supercontinuum (pSC) with the pump + idler sum-frequency mixing (SFM) light [8,9]. The pump pulses from the other port of the 50:50 beam-splitter were compressed by a pair of transmission gratings and then coupled into a 1.5 m photonic crystal fiber (PCF) for generating pSC extending to  $\sim 800$  nm. The spatially overlapped idler and unconverted pump beams exiting the OPO mirror M2 were focused by a concave silver mirror into a 2 mm long PPLN crystal with a grating period of 22  $\mu\text{m}$  to generate pump + idler ( $p+i$ ) SFM light with an average power of 0.5 mW. The beam was then collimated by another concave silver mirror. A dichroic mirror was used to separate the idler from the SFM light at  $\sim 807$  nm. The  $p+i$  SFM light was combined with the pSC in an interferometer before detection by an avalanche photodiode (APD). By adjusting the time delay between the  $p+i$  and pSC arms, a beat signal was detected containing the idler CEO frequencies of both channels.

For an SPOPO, the pump  $f_{\text{CEO}}$  equals the sum of the CEO frequencies of the signal and idler [9]; therefore the idler  $f_{\text{CEO}}$  of either channel can be adjusted by changing the pump  $f_{\text{CEO}}$  of the same channel by varying the drive current of the appropriate laser diode in the mode-locked Yb:KYW laser. Alternatively, the idler  $f_{\text{CEO}}$  of both channels may be adjusted by changing the signal  $f_{\text{CEO}}$  by varying the OPO cavity length. A 10 MHz shift in the idler  $f_{\text{CEO}}$  was measured for an adjustment of 100 mA in the seed laser pump current, and a 50 MHz shift was obtained by moving mirror M3 by 400 nm with a piezoelectric transducer (PZT). For an OPO pumped with two asynchronous pump lasers, the idler  $f_{\text{CEO}}$  of each channel can therefore be independently controlled by using these two mechanisms.

In total, four locking loops were implemented to generate fully stabilized asynchronous mid-IR frequency combs. The repetition frequency of laser 1 was stabilized to a 100 MHz RF reference from a frequency synthesizer by applying a feedback signal to a PZT attached to a cavity mirror in the laser. The repetition frequency of laser 2 was stabilized by locking the repetition-frequency difference,  $\Delta f$ , between itself and laser 1 to an audio frequency reference (e.g., 100 Hz) from an electronic function generator by feeding back the error signal to a PZT attached to a cavity mirror in laser 2 [10]. The idler pulses inherited the repetition-rate stabilities of the two Yb:KYW lasers because the OPO was synchronously pumped. The idler CEO frequencies of channel 1 and channel 2 were locked to 10 and 25 MHz, respectively, using locking loops based on digital phase-frequency detectors (PFDs). As shown in Fig. 1, the output from the APD was divided in two by an RF power splitter. One signal passed through a 10 MHz band-pass filter (BPF) and was mixed in a PFD with a 10 MHz reference derived from a synthesizer. After conditioning in a proportional-integral (PI) amplifier, the PFD output was used to lock the idler  $f_{\text{CEO}}$  of channel 1 by actuating an OPO cavity mirror mounted on a PZT. The other signal was filtered by a 25 MHz BPF, and mixed in a PFD with a 25 MHz reference derived via a divide-by-4 circuit from the 100 MHz signal used for locking  $f_{\text{rep}}$

of channel 1. The PFD output was used to lock the idler  $f_{\text{CEO}}$  of channel 2 by adjusting the pump current for laser 2 after conditioning by another PI amplifier.

Shown in Fig. 2 are the RF spectra of the APD signal when (a) both channels were present and both idler  $f_{\text{CEO}}$  locked, (b) only channel 1 was present and its idler  $f_{\text{CEO}}$  locked, and (c) only channel 2 was present and its idler  $f_{\text{CEO}}$  locked. Figures 2(d) and 2(e) show the RF spectrum of the locked  $f_{\text{CEO}}$  beat signal of channel 2 at different resolution bandwidths (RBWs). The  $-3$  dB bandwidth was measured to be 10 Hz [Fig. 2(e)], which was resolution-limited. A  $-3$  dB bandwidth of 10 Hz was also measured for the locked  $f_{\text{CEO}}$  beat signal of channel 1.

The RF spectrum shown in Fig. 2(a) includes not only  $f_{\text{rep}}$  (100 MHz),  $f_{\text{CEO}}$  (10 and 25 MHz), and  $f_{\text{rep}} - f_{\text{CEO}}$  (75 and 90 MHz), but also other spectral components, which we believe originated from interactions among the signals from the two channels. The signals incident on the APD were the  $p+i$  SFM and pSC from channels 1 and 2, which resulted in the six first-order interactions listed in Table 1. Importantly, only the synchronous interactions between  $p+i$  and pSC light from the same channel are continuous, and contain the idler  $f_{\text{CEO}}$  beats of interest. All other interactions are asynchronous and therefore intermittent, occurring with a period of  $1/\Delta f$ . Higher-order interactions involving the sum or difference frequencies between the listed interactions can exist as well, but they are all asynchronous and intermittent.

To illustrate an asynchronous interaction detected by the APD, we blocked the light from the pSC part and

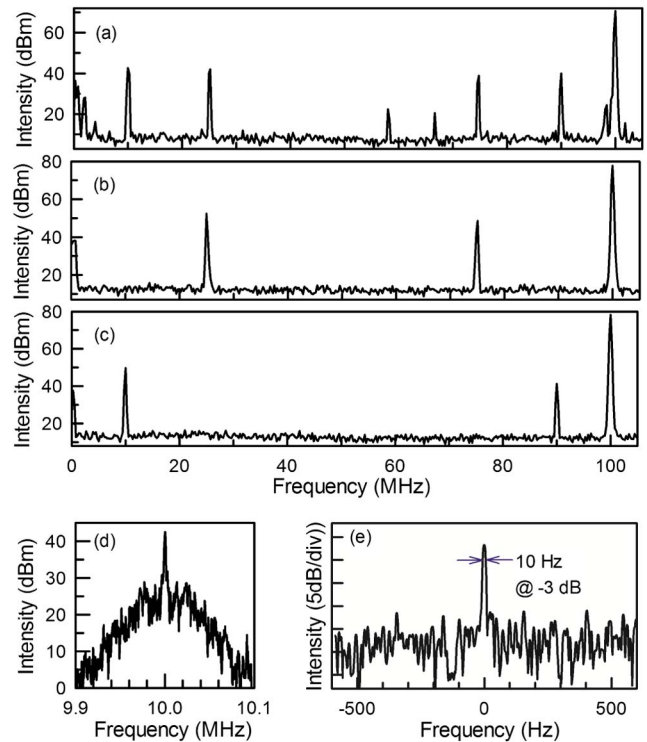


Fig. 2. RF spectra of the APD signal under different conditions, with  $f_{\text{rep}} = 100$  MHz and  $\Delta f = 100$  Hz (see text for details). For (a)–(c) the resolution bandwidth (RBW) and sweep time were, respectively, 100 kHz and 13.5 ms. For (d) and (e), respectively, the RBWs were 1 kHz and 10 Hz, and the sweep times were 258 ms and 12 s.

**Table 1. Interactions Among  $(p+i)_1$ ,  $(p+i)_2$ , pSC1, and pSC2**

Interaction	Spectral Information	Character
$(p+i)_1$ and pSC1	$f_{\text{CEO}}, i, 1$	Continuous
$(p+i)_2$ and pSC2	$f_{\text{CEO}}, i, 2$	Continuous
pSC1 and pSC2	$f_{\text{CEO}}, p, 1 - f_{\text{CEO}}, p, 2$	Intermittent
$(p+i)_1$ and $(p+i)_2$	$f_{\text{CEO}}, (p+i), 1 - f_{\text{CEO}}, (p+i), 2$	Intermittent
$(p+i)_1$ and pSC2	$f_{\text{CEO}}, (p+i), 1 - f_{\text{CEO}}, p, 2$	Intermittent
$(p+i)_2$ and pSC1	$f_{\text{CEO}}, (p+i), 2 - f_{\text{CEO}}, p, 1$	Intermittent

detected only the dual SFM signal. After passing through a 50 MHz RF low-pass filter (LPF), this contains only the interaction between the asynchronous  $(p+i)_1$  and  $(p+i)_2$  channels, and forms a periodic series of interferograms [Fig. 3(a)]. The signal was recorded with a 14 bit data acquisition card at a sampling rate of 100 MHz when  $\Delta f$  was stabilized at 100 Hz, which is confirmed by the 10 ms period ( $1/\Delta f$ ) in Fig. 3(a). A zoomed-in image for one interferogram [Fig. 3(b)] shows that the duty cycle of the intermittent signal is  $\sim 0.0001$ , which can therefore only impose a negligible effect on the stability of the  $f_{\text{CEO}}$  locking because the signals for the synchronous interactions are continuous (duty cycle of 1) and because the integration process in the locking loop averages out small perturbations. Indeed, in the experiments, the existence of asynchronous interactions did not affect the stability of idler  $f_{\text{CEO}}$  locking.

The power spectrum of the Fourier transform (FT) of the interferogram in Fig. 3(b) provides the optical spectrum of the SFM light with a frequency scaling of  $f_{\text{CEO}}/\Delta f$ , and its central frequency indicates the  $f_{\text{CEO}}$  difference between  $(p+i)_1$  and  $(p+i)_2$ . The inset of Fig. 3(b) compares the optical spectrum obtained from the FT (solid line) with the spectra of  $(p+i)_1$  and  $(p+i)_2$  measured with an optical spectrum analyzer (OSA). Absolute wavelength information is not available in this measurement, since the  $f_{\text{CEO}}$  frequencies of  $(p+i)_1$  and  $(p+i)_2$  are not stable; however, mid-IR interferograms using the asynchronous  $f_{\text{CEO}}$ -stabilized idler channels should allow us to extract absolute wavelength information in the mid-IR.

In summary, we have demonstrated dual high-power asynchronous mid-IR frequency combs with fully stabilized repetition rates and CEO frequencies, which are ideally suited for mid-IR coherent frequency comb spectroscopy. The power level we achieved represents a hundred-fold improvement over existing asynchronous mid-IR systems [3,4]. Uniquely, the two comb channels, although pumped by two independent mode-locked pump lasers, share all the components for mid-IR generation and  $f_{\text{CEO}}$  detection. Coupling and interactions between the two channels are negligible, so they behave as two independently controllable frequency comb

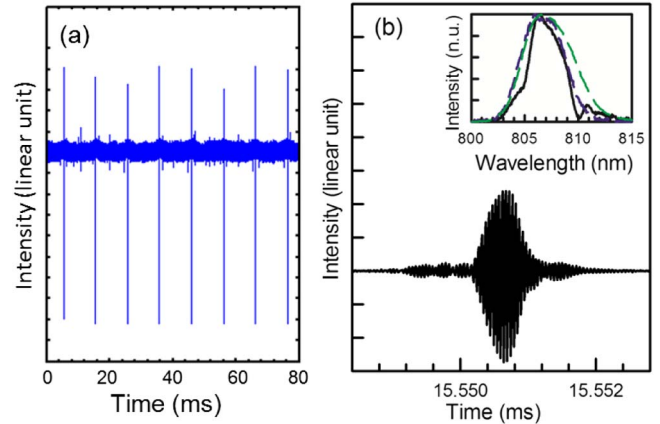


Fig. 3. (a) Detected signal on the APD [after a 50 MHz low-pass filter (LPF)] when only the  $p+i$  SFM outputs from the two channels were present ( $\Delta f = 100$  Hz). (b) Zoomed-in image for one interferogram in (a), and (inset) optical spectrum obtained by Fourier transforming the interferogram (solid line) and comparison to each individual channel measured with an OSA (dashed lines).

systems. This technical route can be readily extended to other spectral ranges, e.g., long-wave mid-IR or the visible region for generating asynchronous dual combs for spectroscopic applications.

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