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# Asynchronous midinfrared ultrafast optical parametric oscillator for dual-comb spectroscopy

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Two asynchronous, broadband 3.3  $\mu\text{m}$  pulse trains with a stabilized repetition-rate difference of up to 5 kHz were generated using an ultrafast optical parametric oscillator. The two oscillation channels, each producing  $\sim 100$  mW average power, ran essentially independently, and weak non-phase-matched sum-frequency mixing between them provided a timing signal that indicated when the asynchronous pulses coincided. The system has immediate applications in incoherent asynchronous optical sampling and, with additional carrier-envelope-offset stabilization, could be applied to coherent dual-frequency-comb spectroscopy. © 2012 Optical Society of America

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Dual-laser pulse trains with slightly different repetition rates are needed for asynchronous optical sampling (AOS) and dual-frequency-comb spectroscopy (DFCS) [1–4]. In these schemes, one pulse train with a repetition rate of  $f_{\text{rep},1}$  (the signal) is sampled by another (the probe) with a repetition rate of  $f_{\text{rep},2} = f_{\text{rep},1} + \Delta f$  ( $\Delta f \ll f_{\text{rep},1}$ ). The relative time delay between the signal and probe pulses increases by  $\Delta f / f_{\text{rep},1}^2$  after every new pair of pulses, and at a given measurement location, the pulses coincide once every  $1/\Delta f$  seconds. The generation of asynchronous pulse trains normally begins with two nearly identical mode-locked lasers, but convenient mode-locked laser sources are not available in the mid-IR, which is one of the spectral regions of most interest for DFCS. Optical parametric oscillators (OPOs) synchronously pumped by near-IR mode-locked lasers can satisfy this requirement by extending the spectral coverage to more than 6  $\mu\text{m}$ .

In recent work we reported a synchronously pumped OPO (SPOPO) producing broadband mid-IR pulses from a cavity containing a long MgO:PPLN crystal [5]. The high intracavity dispersion of this OPO made it insensitive to cavity-length variations, giving oscillation over a 120  $\mu\text{m}$  range (equivalent to an 8 kHz repetition rate detuning) with only 3 nm of signal tuning. This insensitivity can be exploited to allow the OPO to be pumped simultaneously by two lasers with different repetition rates, enabling asynchronous signal/idler pulse trains to be generated with only with slightly different center wavelengths.

In this Letter, we report the first, to our knowledge, SPOPO pumped by a pair of asynchronous pulse trains. The configuration of this asynchronous SPOPO is shown in Fig. 1. Two independent pump sources with identical structure and performance but slightly different repetition rates were combined by a 50:50 plate beam splitter. Each pump source was composed of a fiber amplifier seeded by a mode-locked Yb:KYW laser. 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  $\sim 3$  ps, respectively. The repetition rate of pump Channel 1 was set at 100 MHz, while the

repetition-rate difference,  $\Delta f$ , between the two channels was detected and stabilized to a reference frequency  $\Delta f_{\text{ref}}$  (derived from an electronic function generator) by feeding the amplified error signal back to a piezo-mounted cavity mirror within Yb:KYW laser 2 [6].

The OPO gain medium was a 20 mm long and 1 mm thick 5 mol.% MgO:PPLN crystal with a grating period of 30.49  $\mu\text{m}$ , housed in an aluminum heat sink maintained at 30 °C. In the cavity, the focusing mirrors M1 and M2 (radius of curvature 150 mm) and plane mirror M3 were coated on YAG substrates for high transmission at the pump and idler wavelengths ( $T > 90\%$  at 1020–1110 and 2300–3700 nm) and high reflectivity at the signal wavelength ( $R > 99.8\%$  at 1400–1800 nm). The OPO was singly resonant for the signal, which was extracted through a 10% output coupler (OC). The generated idler and depleted pump passed through M2 and were collimated by a CaF<sub>2</sub> lens, following which an antireflection-coated Ge window was used to isolate the idler pulses prior to characterization. Figure 2 shows the measured signal/idler powers and spectra when the OPO was pumped individually by Channel 1 or Channel 2, or simultaneously by both channels, whose repetition-rate difference was stabilized to 20 Hz.

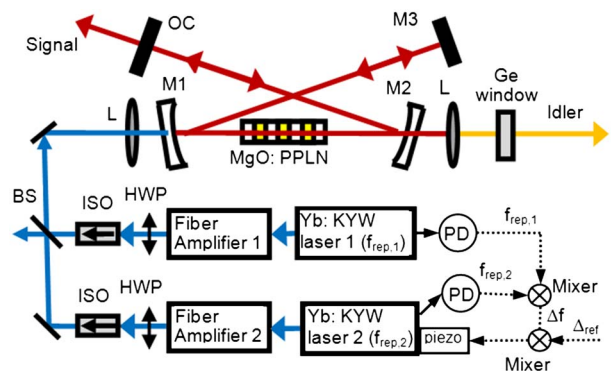


Fig. 1. (Color online) Asynchronous SPOPO layout. HWP, half-wave plate; ISO, isolator; BS, beam splitter; L, lens; OC, output coupler; PD, photodiode.

When pumped only by Channel 1, the threshold pump power was 340 mW. At the full pump power of 1.25 W, the signal (idler) output power was 477 mW (110 mW) and the center wavelength of the signal (idler) pulse was 1543.6 nm (3313 nm), with a 3 dB bandwidth of 2.1 nm (172 nm). The narrow signal bandwidth is due to the phase-matching characteristics of the long MgO:PPLN grating, and consequently the bandwidth of the idler pulses (4.70 THz) closely followed that of the pump (4.82 THz), showing efficient parametric transfer [5].

When pumped only by Channel 2, which was detuned with respect to Channel 1 by  $\Delta f = 20$  Hz, the OPO showed almost identical performance. The center wavelength of the signal (idler) pulse was 1543.5 nm (3322 nm). Normally when the repetition frequency of the pulses pumping an SPOPO is changed, the signal will move to a new center wavelength, so that the cavity round-trip time of the signal pulses still equals the pump-pulse interval. In an SPOPO cavity, which is nearly group-delay dispersion compensated, this effect is typically very pronounced; however, the high group-delay dispersion of our system ( $4270 \text{ fs}^2$ ), which is mainly contributed by the double-passed 20 mm long MgO:PPLN crystal, meant that there was only a 0.1 nm shift of the signal center wavelength. Consequently, the outputs in Channels 1 and 2 can be considered equivalent, with the exception of their repetition frequencies.

When the OPO was pumped simultaneously by both channels, the overall signal/idler output power was measured as the pump power of Channel 1 was varied, keeping Channel 2 pumped at full power. As shown in Figs. 2(a) and 2(b), the measured overall output power was equal to the linear sum of the output powers when individually pumped. Also shown in Figs. 2(c) and 2(d) are the signal and idler spectra when the OPO was pumped simultaneously by both channels at full pump power. These spectra are also nearly identical to the linear sum of the spectra obtained under individual pumping.

The fact that the OPO output when simultaneously pumped is simply a linear summation of the outputs obtained when individually pumped shows that there is no significant nonlinear coupling between the two OPO channels. Further evidence confirming this is shown in Fig. 3. The repetition-rate difference was stabilized to 1 kHz, and the signal pulses produced individually from Channel 1 and Channel 2 were detected by a high-speed InGaAs photodiode and are shown in Figs. 3(a) and 3(b). Because of the repetition-rate difference of 1 kHz, the two signal pulse trains coincided every 1 ms. Shown in Figs. 3(c) and 3(d) are, respectively, the combined signals when the two pulse trains overlap and do not overlap in the time domain. In both cases, the overall signal output is a linear summation of the two individual signal pulse trains. Therefore, each channel oscillates independently of the other, and the OPO with two pumping channels runs like two independent OPOs. This result is due to the instantaneous nature of the parametric gain process.

For an SPOPO with a fixed cavity length, the signal center wavelength can move automatically to adapt to a small change in the pump repetition rate; however, phase-matching considerations mean that the signal

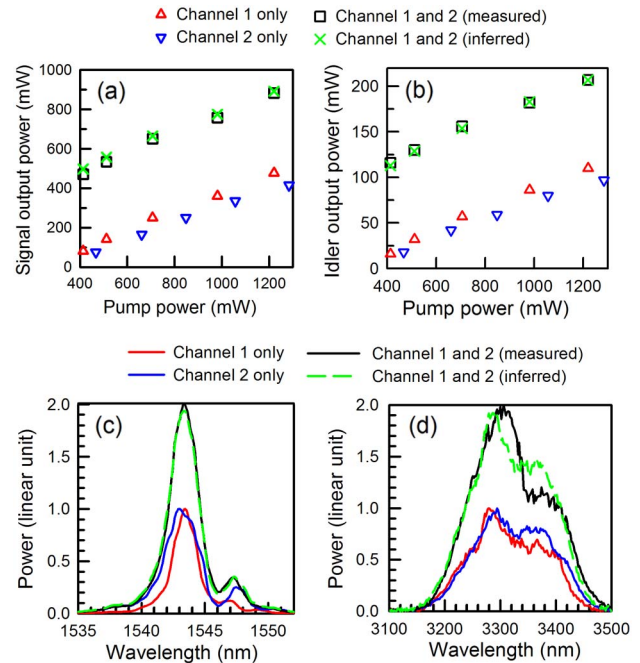


Fig. 2. (Color online) (a) Signal and (b) idler output power from the OPO. The overall signal (idler) output power from Channels 1 and 2 was measured/inferred versus the pump power at Channel 1 while Channel 2 was pumped at full power. (c) Signal and (d) idler output spectra (at full pump power). The inferred total power and spectra for Channels 1 and 2 were obtained by adding those measured for each individual channel ( $\Delta f = 20$  Hz).

tuning is confined to within a finite gain bandwidth. The signal output power falls if its center wavelength shifts away from the peak of this gain curve. In Fig. 4, we show the normalized total signal output power when the OPO was simultaneously pumped by both channels while the repetition-rate difference was increased. For each test, the OPO cavity length was manually adjusted so that the signal output powers from each channel were equal. The data show that at a repetition-rate difference

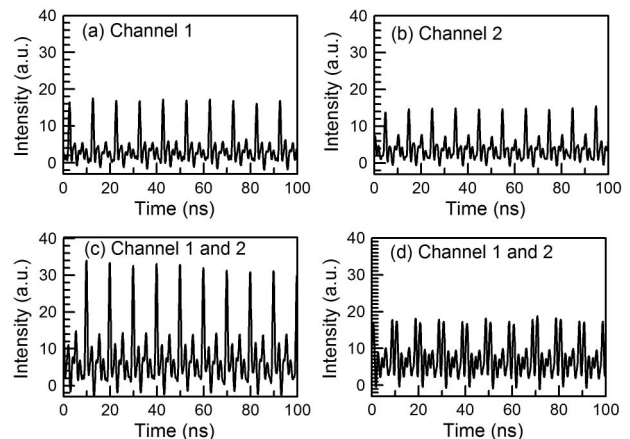


Fig. 3. Signal pulse train from (a) Channel 1 individually, (b) Channel 2 individually, (c) Channels 1 and 2 simultaneously when the pulses from the two pulse trains overlapped, and (d) Channels 1 and 2 simultaneously when the pulses from the two pulse trains did not overlap in the time domain ( $\Delta f = 1$  kHz).

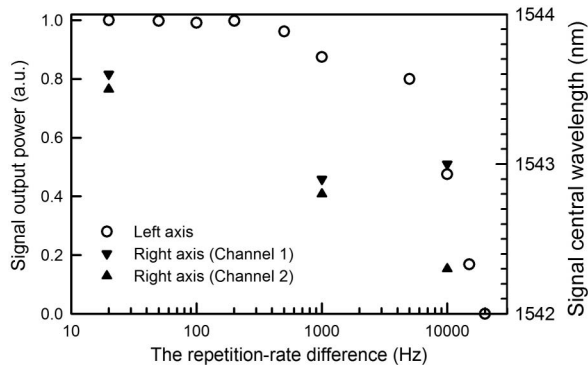


Fig. 4. Dependence on the repetition-rate difference of the normalized total signal output power (circles, left axis) and the center wavelengths of the signal pulses (triangles, right axis).

of  $\sim 5$  kHz, the total signal output power fell to 50% of the maximum.

Also shown in Fig. 4 are the signal center wavelengths of the two channels when the repetition-rate differences were 20 Hz, 1 kHz, and 10 kHz. Even at 10 kHz detuning, the offset in the signal center wavelengths of the two channels was below 1 nm, implying only a 4 nm difference in the center wavelengths of the corresponding idler pulses. With suitable phase stabilization, the asynchronous mid-IR idler pulse trains generated with common center wavelengths can be used for DFCS [3]. For applications requiring asynchronous pulse trains with different center wavelengths, an OPO cavity with smaller group-delay dispersion can be used [7].

It is fundamental to know exactly when the pulses from the asynchronous pulse trains coincide. Red light with a center wavelength at 772 nm was generated by non-phase-matched frequency doubling of the signal pulses and was detected after mirror M3. The intensity of this output was recorded when the OPO was pumped by both channels at a repetition-rate difference of 1 kHz and is shown in Fig. 5. The peaks in Fig. 5 indicate the moment the signal pulses from the two channels overlap, when both the sum frequency (SF) between the two signals and the second harmonic (SH) of each individual signal channel were generated along the MgO:PPLN crystal [8]. The relatively wide pedestals near the peaks arise from the SH of each individual signal channel, which was detected by a photodiode with a rise time much longer than the optical pulse duration, which leads to a higher signal in the way already shown in Fig. 3(c). Beyond the pedestal the interpulse spacing exceeds the rise time of the photodiode and the background levels falls, as previously shown in Fig. 3(d). On an expanded time scale, the peaks repeat every millisecond ( $1/\Delta f$ ), because we have locked the repetition-rate difference between the two channels to 1 kHz. When the signal pulses from the two channels are not temporally overlapped, only the SH of each individual signal channel is generated in the crystal, resulting in the baseline signal in Fig. 5. This parasitic

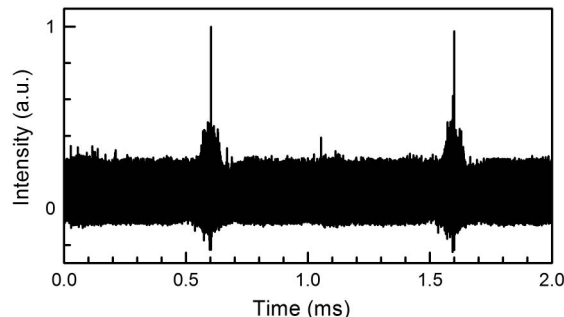


Fig. 5. Non-phase-matched SH/SF signal from the OPO ( $\Delta f = 1$  kHz).

mixing signal is in fact a cross-correlation between the asynchronous OPO signals, similar to that reported in [1], and can be used as a trigger for implementing AOS or DFCS.

In summary, we have demonstrated a single OPO simultaneously pumped by two pump lasers that generates stabilized asynchronous mid-IR pulse trains with nearly identical spectral properties. The OPO cavity acts a spatial mode filter, ensuring that the two pulse trains have identical divergence and perfect spatial overlap, which is critical in applications requiring copropagation over long distances. The system has immediate applications in incoherent AOS, and the low noise of the OPO [5] should make it well suited for this application. Coherent DFCS requires the carrier-envelope offsets of the idler pulses to be stabilized, and this can be readily achieved by heterodyning a pump supercontinuum with the pump-idler sum-frequency mixing light [8]. Carrier-envelope offset control is currently being implemented on the OPO.

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