Pump-tuned deep-infrared femtosecond optical parametric oscillator across 6-7 µm based on $CdSiP_2$

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We report a high-power femtosecond optical parametric oscillator (OPO) at 80 MHz repetition rate, tunable across 6318-7061 nm in the deep-infrared (deep-IR) using pump wavelength tuning. The OPO, based on $CdSiP_2$ (CSP), is synchronously pumped by a commercial Ti:sapphire-pumped femtosecond OPO in the near-IR, enabling rapid static tuning of the CSP OPO with minimal adjustments to the cavity length. The deep-IR CSP OPO provides as much as 32 mW of average idler power at 6808 nm with spectral bandwidth >1000 nm (-10 dB level) across the tuning range. By implementing intracavity dispersion control, near-transform-limited signal pulses of ~100 fs duration with smooth single-peak spectrum are achieved at 1264 nm, corresponding to an idler wavelength at 6440 nm. To the best of our knowledge, this is the first time such practical idler powers in the deep-IR have been generated from a dispersion-compensated CSP femtosecond OPO at sub-100 MHz repetition rate.

OCIS codes: *(190.4360) Nonlinear optics, devices; (190.7110) Ultrafast nonlinear optics; (190.4400) Nonlinear optics, materials; (190.4970) Parametric oscillators and amplifiers.*

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High-power, high-repetition-rate, femtosecond sources in the mid-infrared (mid-IR) are of great demand for variety of applications from material characterization [1] to molecular spectroscopy [2] and human surgery [3]. Such sources also represent a viable route for generating mid-IR frequency combs [4]. Synchronously-pumped optical parametric oscillators (OPOs) based on nonlinear frequency conversion provide a highly effective approach to the generation of high-power widely tunable mid-IR radiation [5]. In spite of the availability of mid-IR nonlinear materials such as $ZnGeP_2$ and OP-GaAs, the newly developed $CdSiP_2$ [6], is rapidly emerging as a highly attractive alternative due to its unique linear and nonlinear properties. Most importantly, the high nonlinearity (d_{eff}~84 pm/V) and wide transparency range in combination with the possibility of phase-matching for pumping at 1 µm using ultrafast solid-state and fiber lasers has been the key to extend wavelength generation to the deep-IR beyond 6 µm [7-10]. At the same time, the large phase-matching bandwidth of CSP, shown in Fig. 1 for a 1-mm-long crystal, can enable rapid and hands-free mid-IR wavelength tuning beyond the traditional cavity delay tuning achieved in femtosecond OPOs [9,10, 11], by exploiting pump tuning. Such broadband pump tuning in the 1 µm wavelength range is now readily available from commercial automated femtosecond OPOs pumped by the well-established Kerr-lens-mode-locked Ti:sapphire laser, providing watt-level average output power levels [12].

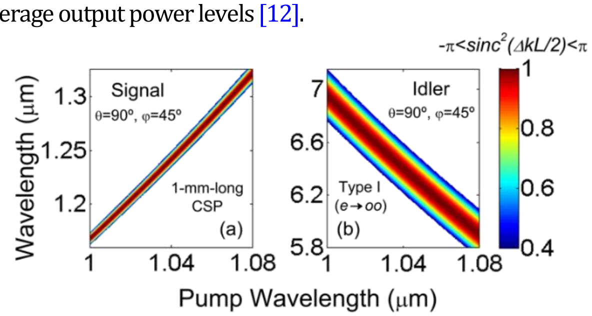

Fig. 1. Parametric gain bandwidth, $-\pi<sinc^2(\Delta kL/2)<\pi$, for type-I ($e\rightarrow oo$) noncritical interaction, provided by a 1-mm-long CSP crystal for the (a) signal, and (b) idler, for pumping from 1 to 1.080 µm.

Here we report the first pump-tuned femtosecond OPO for the deep-IR based on the CSP crystal, tunable across 6318-7061 nm, with -10 dB idler bandwidths >1000 nm across the tuning range. The OPO providing as much as 32 mW of deep-IR average idler power at 6808 nm in near-transform-limited signal pulses of ~100 fs duration with smooth single-peak spectrum.

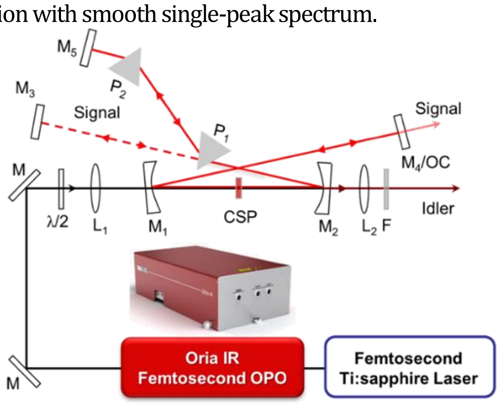

Fig. 2. Schematic of the experimental setup for the pump-tuned CSP femtosecond OPO. λ/2: Half-wave plate, L: Lens, M: Mirrors, OC: Output coupler, P: Prisms, F: Filter. Inset: Oria IR femtosecond OPO.

The schematic of the experimental setup for the pump-tuned deep-IR CSP femtosecond OPO is shown in Fig. 2. The primary pump source is a KLM Ti:sapphire laser (Spectra-Physics, *Mai Tai*) providing up to 2.5 W of average power in ~100 fs pulses at 80 MHz repetition rate, operating at 800 nm. The laser synchronously pumps a commercial fully automated and sealed near- to mid-IR femtosecond OPO (Radiantis, *Oria IR*), shown in the inset of Fig. 2, which provides tunable signal pulses across 990-1550 nm, with >1 W of average power at 1028 nm. However, in the present experiment, we only use the tunable range of 1015-1074 nm. The signal output from the Oria IR is used to synchronously pump a second femtosecond OPO based on CSP to generate the deep-IR radiation beyond 6 µm. The CSP crystal is 1-mm-long with a 4×5 mm^2 aperture, and is cut at θ=90º, ϕ=45º for type-I ($e\rightarrow oo$) noncritical phase-matching. The crystal end-faces are both antireflection (AR)-coated, providing high transmission for the pump (T>99% at 1000-1080 nm), signal (T> 99% over 1160–1240 nm), and idler (T>93% over 6000–7500 nm). The pump beam from the Oria IR is focused to a waist radius of w_0~44 µm at the center of the CSP crystal. The OPO is configured in a standing-wave X-cavity with two plano-concave mirrors, $M_{1\text{-}2}$ (r=150 mm), and two plane mirrors, $M_{3\text{-}4}$. All mirrors are made of 3-mm-thick ZnSe substrate. The mirror coatings provide high transmission at the pump (T>70% at 1020 nm to T>98% at 1080 nm) and **idler (T>95% over 5000–8000 nm),** and high reflectivity for the signal (R>97% over 1200–1230 nm), ensuring singly resonant signal **oscillator (SRO) configuration**. A CaF_2 lens, L_2, is used to collect the idler, while a filter, F, separates the generated deep-IR idler from the residual pump. The total round-trip optical length of the cavity including the CSP crystal is ~3.75 m, corresponding to 80 MHz repetition rate, ensuring synchronization with the Oria IR. In order to characterize the signal pulses from the CSP OPO, we replace one of the plane mirrors with a 5% output coupler (OC). Further, we implement internal dispersion control using a prism pair, $P_{1\text{-}2}$, within the OPO cavity, to achieve transform-limited output pulses.

The average power across the signal tuning range of the Oria IR used to pump the CSP OPO is shown in Fig. 3. The signal power varies from 0.98 W at 1015 nm to 0.68 W at 1074 nm, with a maximum of 1.03 W at 1028 nm. The fully automated Oria IR enables hands-free and continuous tuning across the entire operating wavelength range with smooth spectra, as shown in the inset of Fig. 3. The full-width at half maximum (FWHM) bandwidth of the output signal from the Oria IR varies from 11 nm at 1015 nm to 17 nm at 1074 nm, with a typical measured pulse duration of ~140 fs at 1053 nm. The beam waist radius of ~44 µm results in a peak intensity of ~1.1 GW/cm^2 for a maximum available pump power of ~740 mW at a pump wavelength of 1028 nm, considering the input mirror AR-coating losses. We have not observed any bulk or AR-coating damage while operating the OPO during the experiments.

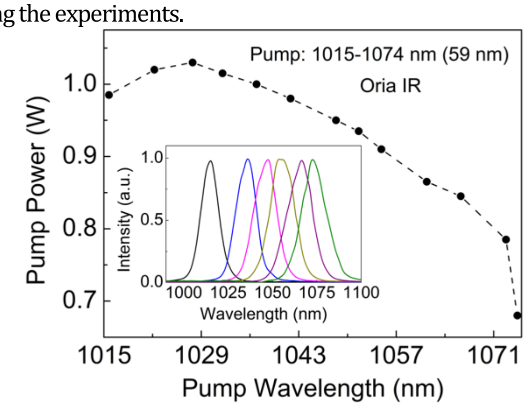

Fig. 3. Power across the tuning range of the Oria IR femtosecond OPO. Inset: Spectra across the tuning range.

In order to characterize the CSP OPO, we initially studied its wavelength tuning performance, which is achieved by varying the output wavelength from the Oria IR, thereby enabling rapid tuning of the CSP OPO with minimal adjustments to the cavity length. The experimentally measured signal spectra across the wavelength range of the pump-tuned CSP OPO are shown in Fig. 4(a), indicating

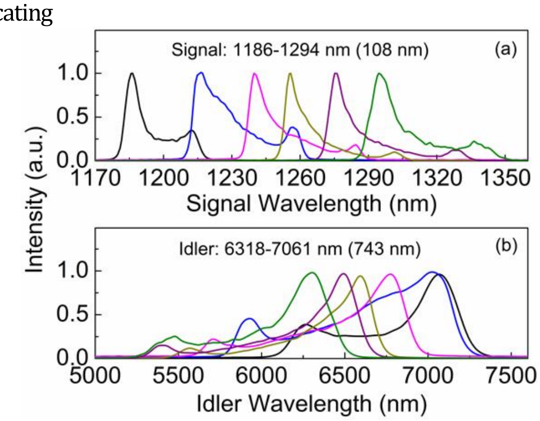

Fig. 4. (a) Measured signal, and (b) reconstructed idler spectra across the tuning range of the pump-tuned CSP femtosecond OPO.

strong self-phase modulation. The reconstructed idler spectra from the Manley-Rowe relations are also shown in Fig. 4(b). It is to be noted that the experimentally measured idler spectra could be different from the reconstructed spectra, particularly due the spectral modulation in the mid-IR region caused by the absorption of various molecular species in the ambient air. However, as evident from Fig. 4(b), while the central pump wavelength from the Oria IR is tuned across 1015-1074 nm, the CSP OPO signal is tunable over 1186-1294 nm (108 nm), corresponding to an idler tuning of 6318-7061 nm (743 nm). The FWHM signal bandwidth varies from ~8 nm at 1186 nm to 12 nm at 1294 nm, with a maximum of ~18 nm (122 cm^{-1}) at 1217 nm. The corresponding idler spectra exhibit an FWHM bandwidth >200 nm across the tuning range with a maximum of 598 nm (122 cm^{-1}) at 7004 nm. At -10 dB level, this corresponds to an idler bandwidth >1000 nm across the tuning range.

The power across the idler tuning range, with the OPO configured as a pure SRO with no output coupling, is shown in Fig. 5. The idler power varies from 18 mW at 6318 nm to 10 mW at 7061 nm, with a maximum of 32 mW at 6808 nm, providing >20 mW of mid-IR idler power and a pump depletion of ~40% over >75% of the tuning range. The typical Oria IR power

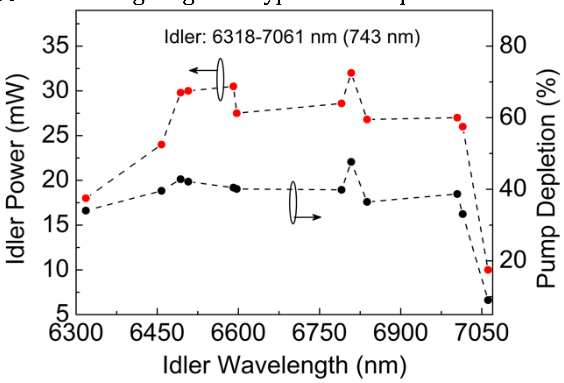

Fig. 5. Deep-IR idler power and pump depletion across the wavelength coverage of the pump-tuned CSP femtosecond OPO.

threshold for the CSP OPO is <0.5 W. Although we operated the CSP OPO as pure SRO for this measurement, we were able to record ~5 mW of signal power through the high reflectors (M_4 and M_5) in each arm, across the tuning range. It is to be noted that the data presented here are not corrected for any losses due to AR coating or absorption in the crystal.

The power scaling results for the pump-tuned CSP OPO are shown in Fig. 6. While operating as pure SRO, we were able to

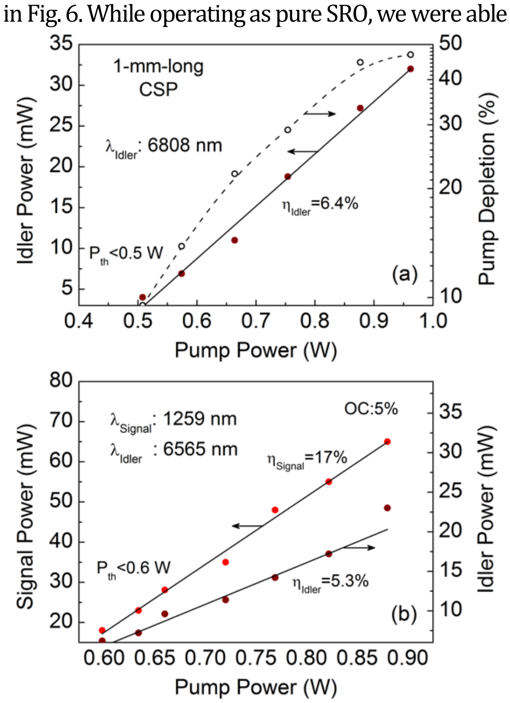

Fig. 6. Power scaling characteristics of the pump-tuned CSP OPO in (a) pure SRO configuration, and (b) using an OC~5%

generate as much as 32 mW of deep-IR idler power at 6808 nm for an input pump power of 0.96 W at 1041 nm, as shown in Fig. 6(a). The corresponding maximum pump depletion is recorded to be ~47%. The OPO threshold in SRO configuration is <0.5 W, while the linear fit to the power scaling data results in an idler slope efficiency of 6.4%. Similar measurements performed at slightly different operating wavelength, by replacing M_4 with a 5% OC are shown in Fig. 6(b), where we were able to extract up to 65 mW of average signal power at 1259 nm together with an idler power of 23 mW at 6565 nm. The corresponding slope efficiencies are estimated to be 17% and 5.3% for the signal and idler, respectively, while the threshold of the OPO with the 5% OC is recorded to be <0.6 W. Although the typical SRO threshold in this wavelength range is also <0.5 W, the marginal increment in the threshold in the presence of the OC is attributed to the high peak pump power from the Oria IR in combination with the large effective nonlinearity of the CSP crystal used in the OPO.

We also performed measurements of the spectral stability of the output signal from the CSP OPO, when operating at a central wavelength of 1259 nm, over a period of 10 min. This result is presented in Fig. 7. The spectra were recorded using a spectrometer with a resolution of ~3 nm. The signal spectrum exhibits self-phase-modulation with a central peak at 1259 nm and the long wavelength tail extending up to 1297 nm. The

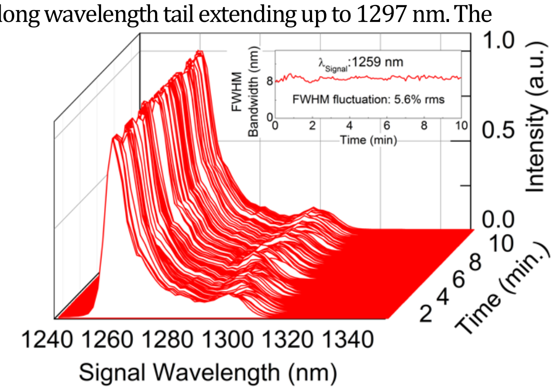

Fig. 7. Spectral stability of the signal extracted from the CSP OPO. Inset: Stability of the FWHM signal bandwidth.

corresponding FWHM signal bandwidth, considering the main peak of the spectrum, is ~8 nm, and exhibits a stability better than 5.6% rms over 10 mins, as shown in the inset of Fig. 7.

The typical intensity autocorrelation trace of the signal pulses extracted from the CSP OPO using a 5% OC is shown in Fig. 8(a). The signal pulses have a duration of ~236 fs (assuming a *sech*^{2} pulse shape). The corresponding self-phase modulated signal spectrum has a FWHM spectral bandwidth of ~9 nm, considering the main peak, when operating at 1253 nm, as shown in the inset of Fig. 8(a). In order to better control the spectral and temporal characteristics of the signal, and hence achieve transform-limited pulses from the CSP OPO, we implemented intracavity dispersion compensation. Using a pair of SF10 prims, P_{1-2}, internal to the CSP OPO, and a high reflector end mirror, M_5, we characterized the signal pulse duration and spectrum. After implementing dispersion compensation, under optimized conditions, we recorded signal pulses with duration of ~100 fs (assuming *sech*^{2} temporal profile), as shown in Fig. 8(b). The corresponding signal spectrum centered at 1264 nm, with a FWHM spectral bandwidth of 19 nm, is shown in the inset of Fig. 8(b), resulting in near-transform-limited pulses with a time-bandwidth product of $\Delta\tau\Delta\nu$~0.36. The signal spectrum is now smooth and single-peak. The corresponding reconstructed idler spectrum in the deep-IR is now also smooth and well-controlled, centered at 6440 nm, with a -10 dB bandwidth of 1193 nm, extending from 5790-6983 nm, as shown in Fig. 9. We, however, note that the idler spectrum may in practice undergo some modulation due to atmospheric absorption. In the presence of intracavity dispersion control, we were able to extract up to 86 mW of signal power together with 22 mW of deep-IR idler power.

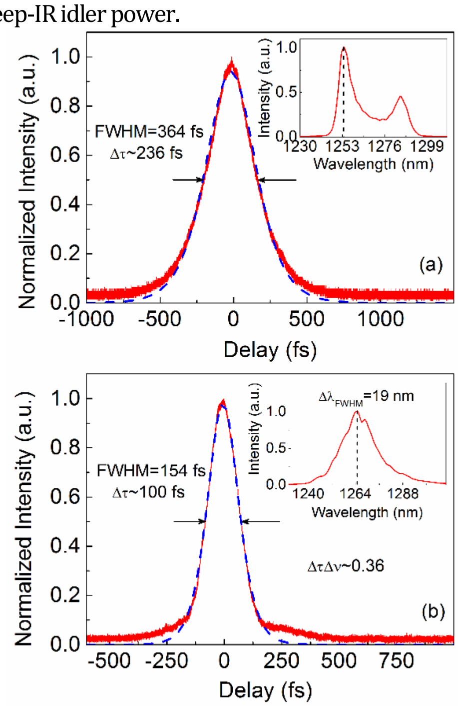

Fig. 8. Temporal profiles of the signal pulses extracted from the CSP femtosecond OPO (a) without, and (b) with dispersion compensation. Dashed blue traces are the *sech²* fit to the experimentally measured profiles. Inset: Corresponding signal spectra.

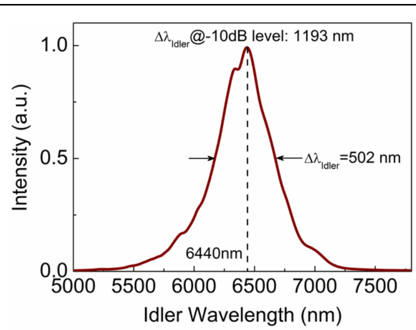

Fig. 9. Reconstructed deep-IR idler spectrum centered at 6440 nm from the dispersion-compensated CSP femtosecond OPO.

In conclusion, we have demonstrated the first pump-tuned femtosecond OPO for the deep-IR based on CSP as the nonlinear gain material, synchronously pumped by a commercial near-IR femtosecond OPO. The CSP OPO is tunable over 6318-7061 nm in the idler and 1186-1294 nm in the signal by rapid automated tuning of the Oria IR output wavelength. The generated idler spectral bandwidths extend up to 1436 nm (-10 dB level) at an operating wavelength of 7004 nm, with >1000 nm over the entire tuning range. The CSP OPO provides up to 32 mW of deep-IR idler power at 6808 nm, with >20 mW over almost the entire tuning range. Further improvements in the output power can be achieved by optimizing the signal output coupling [13]. The signal pulses extracted from the dispersion-compensated CSP OPO cavity are also near-transform-limited, of ~100 fs duration, and with a smooth single-peak spectrum, indicating a similar temporal quality in the mid-IR idler pulses. These results confirm that the exploitation of pump tuning available from commercial femtosecond OPOs in combination with CSP is a highly effective approach for the development of practical femtosecond OPOs in the deep-IR, making the described system a viable and reliable source for many applications including ultrafast spectroscopy and frequency comb generation.

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