Ti:sapphire-pumped deep-infrared femtosecond optical parametric oscillator based on CdSiP₂

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We report a femtosecond optical parametric oscillator (OPO) for the deep-infrared (deep-IR) based on the Kerrlens-mode-locked (KLM) Ti:sapphire laser as the pump source. By deploying a novel cascaded intracavity arrangement comprising a femtosecond OPO based on the nonlinear crystal, CdSiP₂ (CSP), synchronously pumped internal to a MgO:PPLN femtosecond OPO, we have generated broadly tunable radiation across 5958-8117 nm using rapid static cavity delay tuning, with a maximum power of 64 µW at 6791 nm, limited by the absorption in mirror substrates as well as polarizationdependent intracavity losses. The deep-IR idler power exhibits excellent passive stability of better than 1.1% rms over 2 hours, with a spectral bandwidth as large as ~650 nm at ~6800 nm. The demonstrated concept is generic and can be similarly deployed in other operating time-scales and wavelength regions, also using different laser pump sources and nonlinear materials. © 2015 **Optical Society of America**

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Coherent ultrafast laser sources in the mid-infrared (mid-IR) are of interest for a wide range of applications including spectroscopy and trace gas sensing [1,2]. With the scarcity of ultrafast solid-state lasers, synchronously-pumped optical parametric oscillators (OPOs) represent a highly effective approach to the generation of ultrashort pulses in this spectral region [3]. Using the Kerr-lens-mode-locked (KLM) Ti:sapphire laser as the pump source, femtosecond OPOs in various configurations, covering spectral regions from below $\sim 1 \,\mu$ m to beyond $\sim 4 \,\mu$ m, have been extensively developed by exploiting oxide-based birefringent nonlinear crystals such as KTiOPO4, KTiOAsO4, RbTiOAsO4, CsTiOAsO4, and BiB₃O₆ [4,5], as well as quasi-phase-matched (QPM) materials including periodically-poled LiNbO3 (PPLN)

[6]. With the advances in fiber laser technology, femtosecond OPOs pumped by ultrafast Yb-fiber lasers at ~1060 nm, covering spectral regions from \sim 1.4 µm to above \sim 4 µm, have also been successfully realized, addressing new application areas, for example in frequency comb generation [7]. On the other hand, after more than two decades of research and development, the extension of spectral range of femtosecond OPOs significantly beyond ~4 µm remains challenging, due to multi-phonon absorption in oxide-based nonlinear crystals. As an alternative, non-oxide QPM crystals such as orientation-patterned GaAs (OP-GaAs) can be potentially exploited for wavelength generation beyond \sim 4 μ m, but this requires pumping above \sim 2 μ m to avoid two-photon absorption (TPA) in this material. Chalcopyrite crystals such as AgGaSe₂, AgGaS₂, and ZnGeP₂, can generate deep mid-IR radiation up to $\sim 10 \ \mu\text{m}$, but must be pumped well above $\sim 1 \ \mu\text{m}$ to similarly avoid two-photon and residual absorption, and material quality still remains a major practical issue yet to be overcome. The newly discovered chalcopyrite nonlinear crystal, cadmium silicon phosphide, CdSiP₂ (CSP), has drawn special attention in recent years

due to its unique linear and nonlinear optical properties [8]. It is an optically uniaxial crystal with high nonlinear coefficient (d_{eff} ~84 pm/V) and wide transparency (\sim 600 nm to \sim 6.5 µm), which allows pumping near 1 µm without the onset of TPA. Importantly, CSP has the unique capability for parametric generation beyond ~6 µm with direct pumping at ~1 μ m under type I ($e \rightarrow oo$) noncritical phase-matching, (NCPM). By exploiting this scheme in CSP, a number of ultrafast picosecond and femtosecond parametric sources have already been successfully demonstrated using mode-locked solid-state and fiber pump lasers at \sim 1 µm, providing spectral coverage in the \sim 6.1-6.7 µm range [9-14]. These include a single-pass picosecond parametric generator (OPG) at ~6.2 µm pumped by a mode-locked amplified diode-pumped Nd:YVO4 laser at 1064 nm at 100 kHz repetition rate [9], and an OPG tunable over \sim 6.1-6.7 µm pumped by a mode-locked cavity-dumped Nd:YAG laser at 1064 nm with a pulse repetition rate of 5 Hz [10]. A picosecond OPO synchronously pumped by a mode-locked amplified Nd:YAG laser at 1064 nm was also reported, generating idler pulses at ~6.4 µm at 100 MHz [11], while a compact high-energy picosecond OPO using a Nd:YAG oscillator-amplifier system at 1064 nm was demonstrated, providing tunable output across ~6.1-6.6 µm at 450 MHz repetition rate [12]. In the femtosecond time-scale, operation

of an OPO based on CSP synchronously pumped by ~130-fs pulses at 1053 nm from a mode-locked Yb:KYW/Yb:fiber oscillator-amplifier was reported with idler output centered at ~6.2 μ m at 100 MHz [13]. More recently, a high-power femtosecond OPO synchronously pumped by ~560-fs pulses from a Yb:KGW thin-disk solid-state laser system at 1029 nm was reported with tuning across 6.5-7.2 μ m at ~43 MHz repetition rate [14].

At the same time, the exploitation of the KLM Ti:sapphire laser as the pump source for the development of femtosecond OPOs beyond ~4 um offers a number of important merits. The laser, which represents the workhorse of ultrafast technology, still remains the most viable and well-established source of femtosecond pulses, capable of delivering the shortest transform-limited pulse durations and the broadest spectral bandwidths. These features can be of great advantage in also ultimately delivering the shortest pulses and largest spectral bandwidths from OPOs, which are important for applications such as optical frequency comb generation and spectroscopy. Using a 20-fs KLM Ti:sapphire laser, a synchronously-pumped OPO in singlyresonant oscillator (SRO) configuration was previously demonstrated, providing broadband few-cycle pulses across ~2-4 µm in the mid-IR [15]. However, the use of the KLM Ti:sapphire laser in combination with chalcopyrite materials for the development of femtosecond OPOs beyond ~4 µm is fundamentally precluded by TPA at pumping wavelengths below $\sim 1 \,\mu m$. In an effort to overcome this constraint, external cascaded pumping using two OPOs in series, based on CTA and AgGaSe₂, was previously deployed, providing tuning up to $\sim 8 \ \mu m$ [16]. However, such external tandem schemes inherently require high input pump powers, and result in relatively complex architectures involving two independent OPO cavities synchronized in series to one another, and to the pump laser, which can also lead to output instabilities. As such, alternative strategies need to be devised for the development of femtosecond OPOs for the deep-IR based on the KLM Ti:sapphire laser as the pump source, but in more simplified and practical designs. Here we report a novel approach to achieve this goal by demonstrating a femtosecond OPO based on the cascaded concept, but deploying an intracavity pumping arrangement, for the first time to our knowledge. In the new scheme, a secondary femtosecond OPO is synchronously pumped internal to a primary OPO, with the two oscillators sharing a common cavity synchronized to a KLM Ti:sapphire pump laser. By deploying MgO:PPLN as the gain material for the primary OPO, we generate signal at a wavelength >1 μ m, which is then utilized to internally pump a secondary OPO based on CSP, thus avoiding TPA in the crystal. Both OPOs are in SRO configuration, where by taking advantage of the high intracavity signal intensities in the primary OPO, successful operation of the secondary OPO is achieved without the need for a high external pump power. Since the two OPOs also share a common cavity in a composite design, the scheme also results in a simplified system architecture, increased output stability, and rapid static tuning. Although wavelength tuning can be achieved in several ways, such as pump, grating and angle tuning, here we use cavity delay tuning. In this scheme, the CSP femtosecond OPO is rapidly tuned across the entire \sim 5.9-8.1 µm by simple variation of its cavity delay without adjustment of any other parameters. The output exhibits excellent passive power stability better than 1.1% rms over 2 hours.

The schematic of the experimental setup for the intracavity cascaded femtosecond OPO is shown in Fig. 1. The setup consists of two OPOs sharing a common cavity, in which the signal generated from a primary MgO:PPLN femtosecond OPO acts as the pump for a secondary OPO based on CSP. The MgO:PPLN OPO is synchronously pumped by a KLM Ti:sapphire laser providing 900 mW of average power in pulses of 155 fs duration at 76 MHz repetition rate. The laser is operated at a central wavelength of 797 nm and has an output spectrum with a full-width-

at-half-maximum (FWHM) spectral bandwidth of \sim 7 nm. To avoid back-reflections, an isolator is deployed between the laser and the

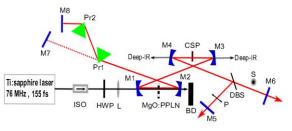


Fig. 1. Experimental setup for the intracavity cascaded OPO. ISO: Faraday isolator, HWP: Half-wave-plate, L: Lens, M1-8: Mirrors, DBS: Dichroic beam-splitter, Pr1-2: Prism pair, BD: Beam dump.

primary OPO and a half-wave plate provides the required polarization for phase-matching in the MgO:PPLN crystal, which is 0.5-mm-long, 3.4-mm-wide and 1-mm-thick, and maintained at 100 °C in an oven. The crystal contains a fan-out grating varying in period from Λ =16 to 23 µm across the 3.4-mm-wide aperture, and the end-faces are antireflection (AR)-coated (R<0.75%) for the signal over 1000-1600 nm and pump (R < 5%) over 720-820 nm, with high transmission (T>85%) for the idler over 1600–3500 nm. The secondary OPO is based on a 0.5-mm-long CSP crystal with a $4 \times 5 \text{ mm}^2$ aperture, cut at θ =90° (φ =45°) for type I ($e \rightarrow oo$) NCPM at room temperature. The crystal faces are AR-coated for high transmission over 1020-1300 nm (T>99%) and 5900-6700 nm (T>95%). The intracavity cascaded OPO is configured as a standing-wave cavity with two internal foci, where the MgO:PPLN and CSP crystals are located. The resonator is formed by a single set of mirrors comprising four concave (r=100 mm), M1-M4, and four plane, M5-M8, mirrors. All mirrors are made of fused silica substrates and are coated for high transmission (T>90%) at 710-840 nm for the KLM Ti:sapphire pump and high reflectivity (*R*>99.8%) over 980-1640 nm for the signal wavelength range in both MgO:PPLN and CSP, ensuring SRO operation in both OPOs. Using a lens of focal length, f=7.5 cm, the pump beam is focused to a waist radius of $w_p \sim 25$ μm in the MgO:PPLN crystal, placed at the center of M1-M2. The cavity design results in a signal beam waist of w_{s1} ~29 µm in both MgO:PPLN as well as the CSP crystal located at the center of M3-M4. The two OPO cavities are separated by a dichroic beam-splitter (DBS) to allow independent control of each resonator length for synchronization with their respective pump pulse trains, since the difference in the refractive index and physical length of the two crystals results in different physical lengths of the cavity for the same round-trip time. The two cavities are then completed by the plane mirrors, M5 and M6, which are mounted on precision translation stages. The DBS is AR-coated for high reflectivity (R>99%) over 1000-1100 nm and high transmission (7>98%) across 1150-1350 nm, thus reflecting the signal from the MgO:PPLN OPO, while transmitting the signal from the CSP OPO. Since the signal waves generated by the two crystals are orthogonally polarized (as indicated by S and P in Fig. 1), one could ideally also use a polarization-dependent beams-splitter to separate the two cavities. At the common end of the cavity, mirror M7 serves as the end mirror for both the OPOs, with M8 serving as the end mirror in the dispersioncompensated cavity. Dispersion compensation is implemented using a pair of intracavity SF11 prisms to achieve the near-transform-limited signal pulses in the MgO:PPLN OPO to pump the CSP OPO.

Before introducing the CSP crystal into the cavity, we first characterized the signal pulses from the dispersion-compensated MgO:PPLN OPO. Using a $2-\mu m$ thin pellicle beam-splitter at Brewster angle located between M4 and DBS, we could extract up to 72 mW of signal power for measurement of pulse duration. A typical interferometric autocorrelation of the MgO:PPLN signal pulses at 1024

nm is shown in Fig. 2. The autocorrelation trace results in a pulse duration of 210 fs (assuming *sech*² pulse shape) with a FWHM spectral bandwidth of 7.3 nm, corresponding to a time-bandwidth product, $\Delta \tau \Delta v \sim 0.44$, close to the transform limit.

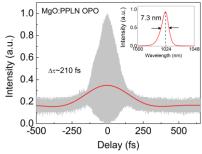


Fig. 2. Interferometric autocorrelation of signal pulses from the MgO:PPLN OPO. Inset: Corresponding spectrum centered at 1024 nm.

After the characterization of the MgO:PPLN OPO signal pulses, we introduced the CSP crystal into the secondary focus of the cavity between M3 and M4. The cavity length of the MgO:PPLN OPO is adjusted to compensate for the additional path length due to the CSP crystal as well as to achieve the required signal spectral characteristics to pump the CSP OPO. The insertion of the CSP resulted in a minimal drop in the MgO:PPLN signal power from 72 mW to 68 mW, confirming the low loss and good transmission of the CSP crystal at 1024 nm. The CSP OPO was synchronized to the MgO:PPLN OPO, and hence the pump laser repetition rate, by fine translation of M6, until oscillation was achieved. Under perfect synchronization, the two signal wavelengths from both OPOs could resonate simultaneously inside the composite cavity, as shown in Fig. 3. The signal wavelength generated by the MgO:PPLN OPO is centered at 1024 nm, while that generated by the CSP OPO is centered at 1194 nm. We also observed generation of strong red light from the CSP crystal due to the non-phase-matched second harmonic generation of the signal at 597 nm, as shown in the inset of Fig. 3. The signal pulses generated in the CSP OPO are affected by the dispersion in the MgO:PPLN crystal, and vice versa. For example, the group delay dispersion experienced by the signal pulses from the CSP OPO at an operating wavelength of 1194 nm is estimated as ~70 fs² considering the ordinary polarization in the MgO:PPLN crystal.

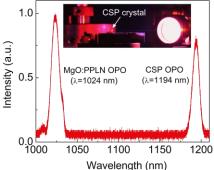


Fig. 3. Simultaneous oscillation of signal wavelengths in MgO:PPLN and CSP OPOs resonant within the composite cavity. Inset: CSP crystal inside the cavity and the bright non-phase-matched red radiation generated by the CSP OPO.

We then systematically characterized the CSP femtosecond OPO. Wavelength tuning in the CSP OPO was achieved only by variation of its cavity length. For a fixed MgO:PPLN signal wavelength of 1024 nm, we were able to tune the CSP OPO signal across 1172-1237 nm by changing the cavity delay over ΔL ~70 µm via translation of M6. The corresponding signal spectra are shown in Fig. 4(a). The FWHM signal

bandwidth varies from 10.5 nm at 1172 nm to 29 nm at 1237 nm. The corresponding reconstructed idler spectra from the Manley-Rowe relations are shown in Fig. 4(b). As can be seen, the central wavelength of the idler from the CSP OPO can be tuned across 5958-8117 nm (over 2159 nm) in the deep-IR, which is to our knowledge the broadest spectral coverage so far achieved with parametric conversion in CSP. Also presented in the inset of Fig. 4(b) is the FWHM idler spectral bandwidth as a function of wavelength, showing a variation from 658 nm at 5958 nm to 508 nm at 8117 nm, with a maximum FWHM bandwidth of 658 nm centered at 6843 nm.

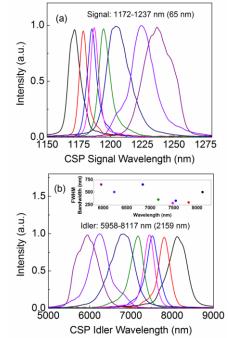


Fig. 4. (a) Signal and (b) reconstructed idler spectra across the CSP OPO tuning range for MgO:PPLN OPO signal wavelength of 1024 nm.

The signal power leakage through the highly reflecting end mirror, M6, and the corresponding wavelength, measured across the tuning range of the CSP OPO, as a function of the cavity delay, for a MgO:PPLN signal wavelength of ~1022 nm, are shown in Fig. 5. As evident, the signal power varies from 6 μ W at 1171 nm for a negative cavity detuning of ΔL =-50 μ m to a maximum of 346 μ W at 1204 nm under perfect synchronization, beyond which it drops down to 44 μ W at 1236 nm for a positive detuning of ΔL =+20 μ m. The corresponding idler wavelengths calculated from energy conservation, vary from 5913 to 8046 nm, as shown in Fig. 5. The data represent the widest tuning together with the longest wavelengths generated from a CSP

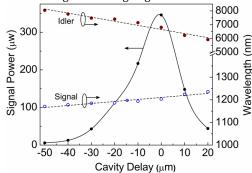


Fig. 5. Signal power and the corresponding CSP OPO signal and idler wavelengths versus the CSP OPO cavity delay.

OPO in any time scale. The ability to achieve such a performance is one of the main advantages of the intracavity cascaded technique, which enables flexible choice of the pump wavelength. Further, we note that the CSP OPO is operated at room temperature with no temperature control for the CSP crystal.

We also characterized the two OPOs with respect to pump depletion. While pumping at the maximum available Ti:sapphire laser power of 900 mW, we recorded a pump depletion of 36% in the MgO:PPLN OPO alone, with the CSP OPO detuned well away from synchronization to cease oscillation. However, with both OPOs perfectly synchronized, we recorded an additional pump depletion of 14%, resulting in a total depletion of 50% in the Ti:sapphire power due to parametric conversion in both OPOs. In order to estimate the change in the intracavity signal power in the MgO:PPLN OPO due to the operation of CSP OPO, we recorded the signal leakage through M5, with and without CSP OPO in oscillation. The result is shown in Fig. 6, where a depletion of ~18% in the intracavity signal power of MgO:PPLN OPO is evident due to the oscillation of the CSP OPO. Even with this relatively low pump depletion, operation of the CSP OPO was sufficiently robust that we could replace the highly reflecting end mirror, M6, with a $\sim 5\%$ output coupler, where we were able to extract

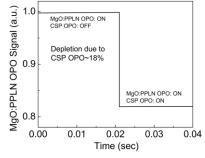


Fig. 6. Depletion of the signal from the MgO:PPLN OPO due the CSP OPO measured using a GaAs photodetector.

up to 9 mW of signal power. Since the intracavity cascaded OPO was configured in a standing wave cavity, the idler output from the CSP OPO could be extracted through both M3 and M4. Owing to the high absorption (>88%) in the 6-mm-thick fused silica substrate of each mirror in the deep-IR wavelength range, we were only able to measure an idler power of 64 μ W. The extracted idler power can be improved by using suitable ZnSe substrates for M3 and M4. The long-term power stability of the deep-IR idler at a wavelength of 6791 nm is shown in Fig. 7, confirming a passive power stability better than 1.1% rms over 2 h. Also shown in the inset of Fig. 7 is the spatial profile of the deep-IR idler beam measured using a pyroelectric camera, indicating a single peak Gaussian distribution.

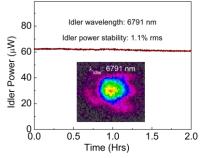


Fig. 7. Long-term power stability of the deep-IR idler from the CSP OPO. Inset: Spatial profile of the deep-IR idler beam.

In conclusion, we have demonstrated a Ti:sapphire-pumped femtosecond OPO for the deep-IR using a novel cascaded intracavity

pumping scheme, for the first time to our knowledge. The OPO provides a record idler spectral coverage across 5958-8117 nm using rapid static cavity delay tuning. The deep-IR idler power of 64 µW is currently limited by absorption in the fused silica mirror substrates as well as the polarization-dependent losses of the intracavity CSP OPO signal at the prism surfaces. Using ZnSe mirror substrates, AR-coated prisms or chirped mirrors for dispersion compensation, and a KLM Ti:sapphire pump laser of higher power, substantial improvements in the deep-IR output power are expected using the demonstrated technique. The composite cavity design results in excellent long-term passive power stability better than 1.1% rms over 2 hours, with a FWHM spectral bandwidth as large as large as 658 nm at 6843 nm. The intracavity cascaded technique is universal and can be deployed in other time-scales and wavelength regions, and using other laser pump sources and nonlinear materials. In particular, the combination of the KLM Ti:sapphire pump laser with technologically mature MgO:PPLN opens up new avenues to exploit the technique for pumping other infrared nonlinear materials such as AgGaSe2, ZnGeP2 and OP-GaAs for deep-IR wavelength generation beyond $\sim 4 \,\mu m$.

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