$CdSiP_2$ optical parametric generator

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ABSTRACT

We report efficient generation of picosecond pulses in the near- and mid-infrared in the new nonlinear material $CdSiP_2$ pumped at 1.064 μm by an amplified mode-locked Nd:YVO_4 laser at 100 kHz repetition rate. Using single-pass optical parametric generation in 8-mm-long crystal cut for type I (*e→oo*) noncritical phase-matching, an average idler power of 154 mW at 6.204 μm together with 1.16 W of signal at 1.282 μm has been obtained for 6.1 W of pump at photon conversion efficiencies of 15% and 23%, respectively. Signal pulse durations of 6.36 ps are measured for 9 ps pump pulses, with both signal and idler beams in near-Gaussian spatial profile.

Keywords: Nonlinear optics, nonlinear materials, parametric oscillators and amplifiers, ultrafast nonlinear optics

1. INTRODUCTION

In the absence of widely available solid-state lasers in the mid-infrared (mid-IR), optical parametric down-conversion has been established as an effective technique for the generation of coherent radiation in this spectral range. Through the exploitation of oxide-based birefringent materials such as $LiNbO_3$, $KTiOAsO_4$ and $RbTiOAsO_4$, and periodically-poled crystals such as PPLN and PPRTA, spectral regions up to ~5 μm can be accessed, but the onset of multiphonon absorption sets a practical upper limit of ~4 μm for wavelength generation in such materials. Chalcogenide nonlinear crystals with transparency in the mid-IR, such as CdSe and $AgGaSe_2$, can provide coherent light at longer wavelengths, but their low bandgap energy precludes pumping near ~1 μm due to two-photon absorption (TPA), thus preventing the use of practical solid-state Nd-based pump lasers. Other chalcogenide materials with larger bandgap, such as $AgGaS_2$, may be pumped near ~1 μm without the onset of TPA, but poor thermomechancial properties including low thermal conductivity, anisotropy of thermal expansion, and low damage threshold prevent practical device implementation. As such, exploitation of many chalcogenide crystals requires long-wavelength laser pump sources with limited availability near ~2 μm, or the deployment of cascaded pumping schemes with the associated complexities. It would, thus, be imperative to explore more viable alternatives for practical generation of mid-IR radiation beyond ~4 μm using direct pumping with Nd-based solid-state lasers near ~1 μm.

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2. CADMIUM SILICON PHOSPHIDE

The nonlinear crystal, cadmium silicon phosphide, $CdSiP_2$ (CSP),[1] is a recently discovered optical material which offers unique linear and nonlinear properties for parametric down-conversion into the mid-IR. It is a negative uniaxial chalcopyrite compound with a transparency above ~6.5 μm, which possesses noncritical phase-matching (NCPM) capability with a maximum effective nonlinear coefficient as high as $d_{eff}=d_{36}=84.5$ pm/V.[2] For pumping near 1 μm, CSP has been shown to outperform other mid-IR materials in almost every respect.[3] Importantly, CSP has a band-gap well below 1 μm, which permits pumping at 1.064 μm, and under type I ($e \rightarrow oo$) parametric generation with NCPM can provide an idler wavelength near 6.4 μm, a spectral range of great interest for medical applications.[4] In earlier studies, the potential of CSP for the generation of mid-IR radiation using direct pumping at 1.064 μm was demonstrated.[5-7] Using a Q-switched Nd:YAG laser, operation of a CSP optical parametric oscillator (OPO) providing idler pulses with 470 μJ of energy at 6.2 μm at 10 Hz repetition rate was achieved.[5] Soon after, using a pulsed mode-locked picosecond Nd:YAG laser in an oscillator-amplifier format, a synchronously-pumped OPO based on CSP, providing idler pulses near 6.4 μm with an energy of 2.8 μJ at 100 MHz, in a train of 2 μs macropulses at a 25 Hz repetition rate was reported.[6] More recently, a sub-nanosecond OPO based on CSP providing idler energy of 24 μJ at 6.125 μm at an average power of 24 mW was demonstrated.[7] Here, we report efficient generation of picosecond pulses in near- and mid-IR in CSP at a repetition rate as high as 100 kHz using single-pass optical parametric generation (OPG) pumped by a mode-locked $Nd:YVO_4$ laser at 1.064 μm.[8] We demonstrate an average signal power of 1.16 W at 1.282 μm and idler power of 154 mW at 6.204 μm for 6.1 W of pump.

2. EXPERIMENTAL

A schematic of the experimental setup is shown in Fig. 1. The pump source is a commercial mode-locked $Nd:YVO_4$ laser at 1.0642 μm (Lumera Laser GmbH, Hyper 50) in an oscillator-amplifier arrangement. It can deliver up to 40 W of average power at 100 kHz, corresponding to an energy of 400 μJ per pulse. The output beam has a diameter of 5 mm and the pulses are transform-limited with durations of 8.7 ps, implying a spectral bandwidth of ~0.2 nm (assuming Gaussian pulse shape). The beam has a quality factor M^2~1.1 and the output power has excellent stability of <0.5% RMS over 13 hours. Using a telescope consisting of uncoated fused silica lenses, the pump beam is collimated to a ~500 μm diameter before the CSP crystal. The pump power and polarization are controlled using two half-wave plates and a polarizing beam-splitter cube. Pumping is single-pass.

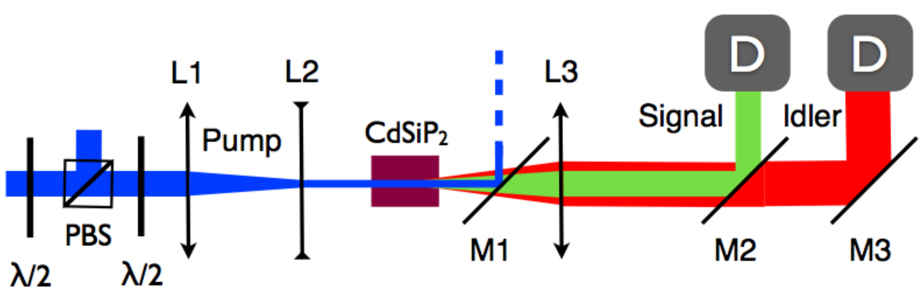


Fig. 1. Experimental setup for single-pass optical parametric generation in CSP pumped at 1.064 μm.
λ/2: half-wave plate, PBS: polarizing beam-splitter, L: lens, M: mirror, D: diagnostics.

The CSP crystal was grown from a stoichiometric melt by the horizontal gradient freeze technique.[9] It was cut at θ=90° (φ=45°) for type I ($e \rightarrow oo$) interaction under NCPM with a length of 8 mm and an aperture of 6.75 mm X 6 mm (along the *c*-axis). The residual loss of the crystal was measured to be 0.198 cm^{-1} for the pump at 1.064 μm, 0.114 cm^{-1} for the signal near 1.3 μm, and 0.014 cm^{-1} for the idler near 6.2 μm. Both crystal faces were antireflection (AR)-coated for the three wavelengths with an eight-layer coating, providing an average reflectivity per surface of ~0.35% at 1064 nm,

~0.4% at 1.275 μm, and ~0.5% at 6.2 μm. The overall single-pass transmission of the AR-coated sample was 83% at 1.064 μm.

For characterization of OPG output, the pump light after transmission through the crystal was rejected using a 1-mm-thick CaF_2 plane dichroic mirror (M1) with high reflectivity (R>99%) at 1.064 μm and transmitting at the signal and idler (T=60% at 1.28 μm, T>90% over 6-7 μm). An uncoated CaF_2 lens, L3 (f=75 mm), was then used to collimate the generated signal beam, which was separated from the idler using a ZnSe mirror (M2) with high transmission at the idler (T>95% over 6-7 μm). For the study of idler, L3 was replaced by another uncoated CaF_2 lens with f=50 mm, since the divergence of the idler beam from the OPG was stronger than that of the signal. The idler transmitted through M2 was then finally reflected by a gold-coated mirror (M3) for characterization. In measurements of power and efficiency, all data were corrected for the transmission and reflection losses through the substrates, uncoated surfaces and mirrors.

2.1 Output Power, energy, and efficiency

Using this setup, by gradual increase of pump power, we observed OPG threshold at an average pump power of 1.1 W at the input to the CSP crystal. This corresponds to a pump pulse energy of 11 μJ at 100 kHz, and pumping intensity of 0.62 GW/cm^2. By further increasing the pump to 6.1 W, we were able to generate an average signal power of 1.16 W at 1.282 μm, with corresponding pulse energy of 11.6 μJ. This represents a power conversion efficiency of ~19% and a photon conversion efficiency of ~23% from the pump to signal. At 6.1 W of pump power, we measured an average idler power of 154 mW at 6.204 μm, corresponding to a pulse energy of 1.54 μJ. Therefore, the power conversion efficiency from the pump to the idler was ~2.5%, with a photon conversion efficiency as much as ~15%. The lower photon conversion efficiency into the idler is attributed to water absorption in the mid-IR, although additional losses in mirror coatings and substrates can not be ruled out. Beyond 6.1 W of input pump power, we observed the onset of lensing in the CSP crystal, with the generated signal and idler beams undergoing strong focusing.

In order to ascertain the origin of this lensing effect, we chopped the pump beam using a mechanical wheel and performed power scaling measurements of OPG output. The results are shown in Fig. 2, where the generated signal pulse energy is plotted as a function of pump pulse energy at the input to the crystal, in the absence of chopping (I), and when the pump is chopped at 100 Hz with duty cycles of 50% (II) and 5% (III). With the pump light unchopped, the signal pulse energy increases linearly for pump pulse energies up to ~40 μJ, beyond which there is evidence of saturation. Increasing the pump pulse energy further, the signal pulse energy rises to 11.6 μJ for 61 μJ of pump pulse energy, at which point we observe focusing of the signal and idler beams. When the pump is chopped, similar linear behavior prevails at low pump pulse energies up to ~40 μJ, but the saturation effect is progressively diminished by reducing the duty cycle. Moreover, under chopped conditions, we no longer observe focusing of the output beams at pump energies above 61 μJ. By further increasing the pump energy, we obtain a signal pulse energy of 14.7 μJ for 80.9 μJ of pump at ~18% conversion efficiency with 50% duty cycle, and 17 μJ for 77.8 μJ of pump at ~22% conversion efficiency with 5% duty cycle. However, as evident, we were not able to completely overcome saturation at higher pump energies even at the highest duty cycle. The results clearly confirm the origin of the saturation and lensing effect as thermal. Given the high repetition rate of the pump laser, saturation in output energy occurs as a result of heating of the crystal, leading to thermal dephasing at higher average powers. At the same time, focusing of the output beams occurs due to the strong thermal lens in the crystal at higher pump powers, and not higher-order intensity-dependent nonlinear optical process such as the Kerr effect. We attribute the thermal effect mainly to the residual absorption of the crystal at the pump wavelength, with some contribution from signal absorption. However, since this absorption is not intrinsic, we expect the growth of CSP samples of higher quality will lead to major reductions in saturation and thermal lensing, allowing significant increase in output power and energy at higher pump powers without chopping. In addition, with the availability of crystals of larger aperture and longer interaction length, we expect substantial increases in the signal and idler power to multiwatt and watt level, respectively, and higher conversion efficiencies, by increasing the available pump power to 40 W, while minimizing saturation and thermal lensing. In the present setup, the available pump power was limited to 10 W, with ~8 W available at the CSP crystal. At the highest pumping intensity of ~4.5 GW/cm^2 used, we observed no sign of optical damage to the CSP crystal or the coatings

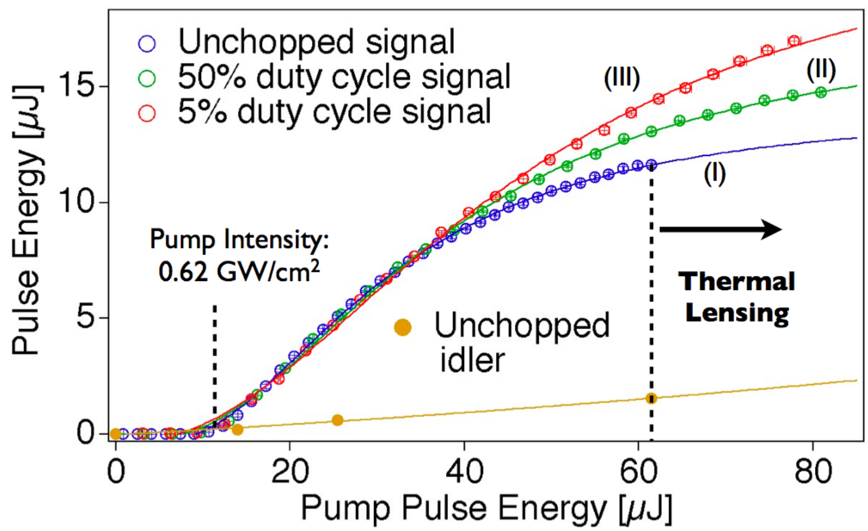
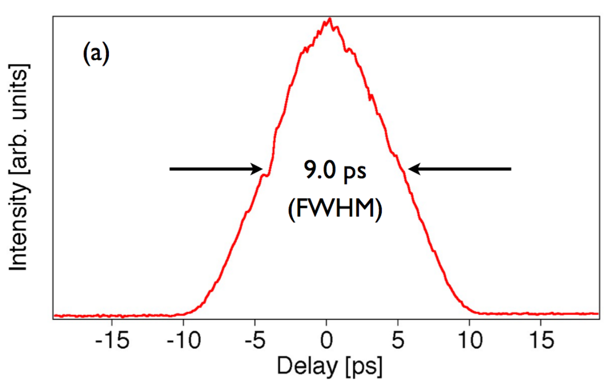


Fig. 2. Output signal pulse energy versus pump pulse energy at the input to the CSP crystal.
(I): Unchopped pump beam; (II), (III): Chopped pump beam. Also shown is the idler pulse energy versus pump pulse energy.

2.2 Temporal and spectral characteristics

We performed temporal measurements of the generated signal pulses using a FROG setup based on a 100 μm BBO crystal, where the device was used simply as an autocorrelator. The output spectra were acquired using a Fourier transform spectrometer equipped with an InGaAs detector for the signal and a HgCdTe detector for the idler. To avoid damage to the detector, the measurements were performed at 2.54 W of average pump power, resulting in 450 mW of signal and 62 mW of idler. Figure 3(a) and 3(b) show the obtained autocorrelation profile of the signal pulse and the corresponding spectrum, respectively. The FWHM of the autocorrelation trace is 9.0 ps, leading to a signal pulse duration of 6.36 ps (assuming Gaussian pulse shape), and the spectrum is centred 1.282 μm with a FWHM bandwidth of 8.5 nm, resulting in a time-bandwidth product of 9.3. Using the Sellmeier equations [2], the calculated pump-signal group velocity mismatch (GVM) is ~231 fs/mm, resulting in a temporal walkoff of ~1.85 ps for the 8-mm crystal. The corresponding pump-idler GVM is ~843 fs/mm, with the signal-idler having a GVM value of ~612 fs/mm.



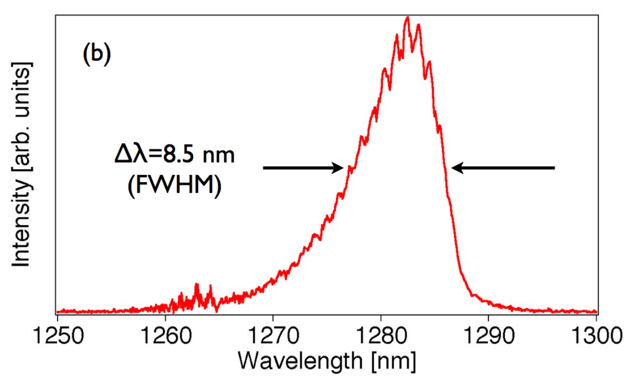


Fig. 3. (a) Intensity autocorrelation, and (b) spectrum of the output signal pulses at 1.282 μm. Input pump power is 2.54 W.

The idler spectrum, shown in Fig. 4, is centred at 6.204 μm, and has a FWHM bandwidth of 122 nm. The dips in the spectrum correspond to absorption lines of water, as verified by the HITRAN molecular database. The signal and idler peak wavelengths of 1.282 μm and 6.204 μm are in close agreement with the calculated values of 1.286 μm and 6.180 μm for a pump wavelength of 1.0642 μm based on the Sellmeier equations [2].

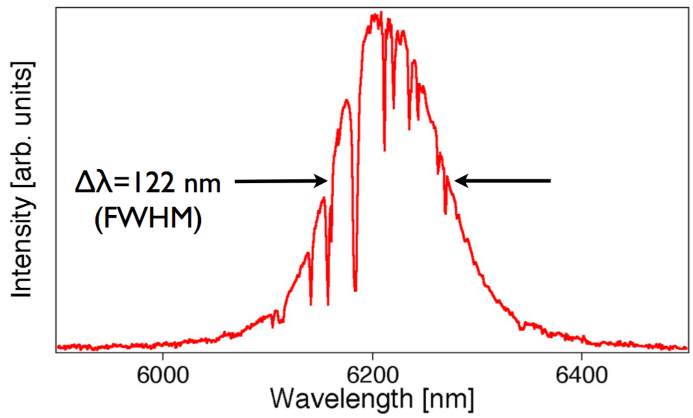


Fig. 4. Measured idler spectrum centered at 6.204 μm. The sharp features correspond to water absorption lines. Input pump power is 2.54 W.

2.3 Spatial beam quality

We also characterized the spatial beam profile of the generated signal and idler using pyroelectric knife-edge scanner. In order to avoid damage to the profiler, the measurements were performed at an average pump power of 2.54 W, resulting in 450 mW of signal and 62 mW of idler. The signal profile was obtained using a CaF_2 lens of focal length *f*=75 mm, providing a collimated beam at the profiler. For the idler beam, we used a CaF_2 leans of *f*=50 mm, but due the larger divergence, the arrangement resulted in the focusing of the idler at the profiler. The obtained beam profiles for the signal and idler are shown in Fig. 5(a) and 5(b). The signal and idler profile have relatively uniform, near-Gaussian energy distribution. The signal has a diameter of 5600μm x 5500μm (FWHM) and the idler has a diameter of 1100μm x 1000μm (FWHM), both with >90% circularity.

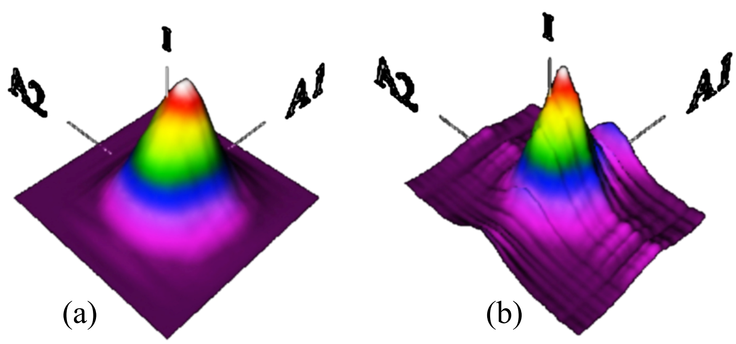


Fig. 5. Spatial profiles of (a) signal beam at 1.282 μm, and (b) idler beam at 6.204 μm. Input pump power is 2.54 W.

3. CONCLUSIONS

In conclusion, we have demonstrated efficient and practical generation of near- and mid-infrared radiation at1.282 μm in the signal and 6.204 μm in the idler using direct single-pass parametric generation in the nonlinear crystal CSP, pumped at 1.064 μm and at a repeititon rate as high as 100 kHz. By using an 8-mm-long crystal cut for type I (*e→oo*) noncritical phase-matching, we have generated an average idler power of 154 mW together with 1.16 W at 1.282 μ of signal for 6.1 W of pump at photon conversion efficiencies of 15% and 23%, respectively. We have obtained signal pulse durations of 6.36 ps for 9 ps pump pulses, with both signal and idler beams in near-Gaussian spatial profile. The limit to the generated signal and idler power is currently set by lensing in the CSP crystal at pump powers above 6.1 W, where we have verified the origin of the effect as thermal. With the availability of crystals of higher optical quality, larger aperture and longer interaction length, we expect substantial increases in the signal and idler power to multiwatt and watt level, respectively, and higher conversion efficiencies, by increasing the available pump power to 40 W, while minimizing saturation and thermal lensing.

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