High-energy, 450 MHz, $CdSiP_2$ picosecond optical parametric oscillator near 6.3 μm for biomedical applications

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ABSTRACT

We report a compact, efficient, high-energy and high-repetition-rate mid-infrared picosecond optical parametric oscillator (OPO) based on a new nonlinear material $CdSiP_2$. The OPO is synchronously pumped by a master-oscillator power-amplifier system at 1064.1 nm, providing 1-µs-long macro-pulses constituting 8.6 ps micro-pulses at 450 MHz, and can be tuned over 486 nm across 6091-6577 nm, covering the technologically important wavelength range for surgical applications. Using a compact cavity (~30 cm) and a $CdSiP_2$ crystal, idler macro-pulse energy as high as 1.5 mJ has been obtained at 6275 nm, for an input energy of 30 mJ. The extracted signal pulses have durations of 10.6 ps.

Keywords: Nonlinear optics, Optical parametric oscillators, Cadium silicon phosphide, High energy lasers.

1. INTRODUCTION

The mid-infrared (mid-IR) window of the optical spectrum has a wealth of information pertaining to many molecules of importance involved in a variety of applications, from surgery[1] to explosive detection.[2] In particular, mid-IR radiation in 5900-6600 nm wavelength range, constituting the absorption bands of amide I (6000 nm), water (6100 nm) and amide II (6450 nm) are of interest for human surgery, because of the simultaneous absorption by both proteins and water. Research in the past has shown that the choice of these wavelengths minimizes the collateral damage, while maintaining an effective ablation rate of the tissue, enabling minimally invasive human surgery.[1] In the absence of conventional lasers at these wavelengths, the free electron laser has so far been shown to be the only source capable of delivering sufficient energy for such applications.[3] But high complexity, large size, and high cost preclude its use in practical applications. Optical parametric oscillators (OPOs) are now well recognized as powerful and viable solid-state sources of coherent radiation, providing access to mid-IR spectral region with wide tunability. However, mid-IR OPOs, pumped at 1 µm have relied mainly on the well-established oxide-based nonlinear materials such as $LiNbO_3$, practically limiting the spectral coverage of these devices to 4 µm, imposed by the onset of absorption.[4] Alternatively, OPOs based on chalcopyrite crystals such as AgGaSe and the most developed $ZnGeP_2$ (ZGP) can generate mid-IR radiation up to 10 µm, but must be pumped above 1 µm to avoid two-photon and residual absorption. As a result, they are often pumped in tandem, typically by another OPO, resulting in complex architectures.[4] Hence, it is imperative to explore viable alternative materials for generation of mid-IR radiation beyond 4 µm pumped by the well-established Nd-based lasers at 1064 nm. The quest for such a nonlinear material recently led to the development of cadmium silicon phosphide, $CdSiP_2$ (CSP).[5] Its large band gap, high optical quality, improved thermal and optical properties with high effective nonlinear coefficient (d_{eff} ~ 84.5 pm/V), and noncritical phase-matching (NCPM), make it an attractive nonlinear material candidate for the generation of mid-IR wavelengths in the 6-6.5 µm range.[6]

Earlier reports on mid-IR OPOs based on CSP pumped at 1064 nm include a low-repetition-rate (10 Hz) nanosecond oscillator with idler energies up to 0.47 mJ at 6.2 µm,[7] and a 0.24 mJ, 1 kHz, sub-nanosecond OPO with idler tuning around 6.1 µm.[8] Also, mid-IR picosecond pulses near 6.4 µm with an output energy of 0.56 mJ were generated by synchronous pumping an OPO with a nonlinear mirror mode-locked Nd:YAG laser in non-collinear geometry.[9] Recently, single-pass parametric generation in CSP was also reported, pumped at 1064 nm, providing picosecond pulses with energy of 1.54 µJ at 6.2 µm at a repetition rate of 100 kHz.[10]

Here we report a compact, 450 MHz repetition rate OPO based on CSP, synchronously pumped by a master oscillator power amplifier (MOPA) system at 1064.1 nm, generating an idler energy as high as 1.5 mJ at 6246 nm with a photon conversion efficiency of 29.5%. The OPO is tunable over 486 nm, with more than 1.2 mJ over >68% of the tuning range in good beam quality. To our knowledge, this is the first high-repetition-rate picosecond OPO based on CSP, synchronously pumped at 1064 nm in collinear geometry.[11]

2. PROPERTIES OF CSP

The new mid-IR nonlinear crystal, CSP, was discovered only recently. It is a chalcopyrite semiconductor belonging to the space group $\bar{4}2m$, with practical transparency extending from 1 to 6.5 µm. Various optical, thermal and mechanical properties of CSP are tabulated in Table-1. CSP is a very interesting nonlinear material, because it possesses most of the attractive properties as ZGP, a crystal which is relatively mature for mid-IR generation. CSP is also uniaxial, but unlike ZGP has negative birefringence. Uniquely and importantly, the linear and nonlinear optical properties of CSP permit parametric generation under NCPM with maximum effective nonlinearity, providing mid-IR idler radiation near 6 µm when pumping at 1064 nm. The possibility of pumping CSP at the well-established Nd:YAG wavelength of 1064 nm to generate mid-IR radiation at mid-IR wavelengths as far as 6 µm is one of the most important feature of this new nonlinear material. The ordinary and extraordinary refractive indices of CSP are given by the Sellmeier equations,[5,12]

Table 1. Properties of cadmium silicon phosphide (CSP) nonlinear crystal.

Transparency	1 – 6.5 µm
Birefringence (ne-no)	-0.051 @ 4 µm
Bandgap	2.2 - 2.45 eV (564 – 506 nm)
Nonlinear coefficient	d_{14}= 53 pm/V , d_{36}= 84.5 pm/V @ 4.56 µm
Thermal conductivity	13.6 W /m-K
Specific heat	446 J/Kg/K
Melting point	1133 ºC
Thermal diffusivity	7.69 mm^2/s
Thermal expansion	-2.8 x 10-6 K^{-1} (perpendicular to c-axis) 10.2 x 10-6 K^{-1} (parallel to c-axis)
Knoop´s hardness	930 Kg/mm^2
Micro-hardness	9.3 GPa

$$n_e^2 = 3.0811 + \frac{6.2791 \times \lambda^2}{\lambda^2 - 0.10452} - 0.0034888 \times \lambda^2$$

(1)

$$n_o^2 = 3.4343 + \frac{5.6137 \times \lambda^2}{\lambda^2 - 0.11609} - 0.0034264 \times \lambda^2$$

where the equations can be used to calculate the phase-matching conditions for type-I (*e*→*oo*) OPO pumped at 1064 nm. The results are shown in Fig. 1(a), indicating that CSP can be pumped at 1.064 µm by solid-state lasers to generate output beyond 6 µm in a single OPO stage, without the need for any tandem OPO stages.

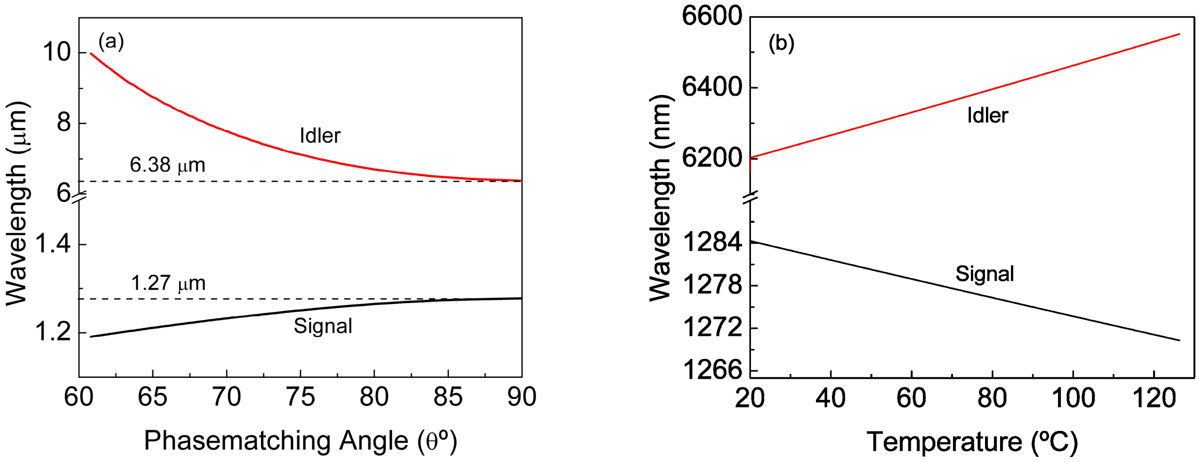

Figure. 1. (a) Angle tuning, and (b) temperature tuning curves for a CSP OPO pumped at 1064 nm.

Further, CSP also has a small, but finite thermal dependence of the refractive indices, enabling temperature tuning. Thermal sensitivity of refractive indices of CSP is given by the temperature-dependent Sellmeier equation,[13]

$$n^2 = A + \frac{B \times \lambda^2}{\lambda^2 - D} - C \times \lambda^2, \qquad (2)$$

where

	n_o	n_e
A	3.0449 + 1.214 × 10−4T (K)	3.3978 + 1.224 × 10−4T (K)
B	6.1164 + 5.459 × 10−4T (K)	5.4297 + 6.174 × 10−4T (K)
C	0.0034888	0.0034888
D	0.10452	0.11609

Figure 1(b) shows the theoretical temperature tuning curves of a CSP OPO pumped at 1064 nm, calculated using the temperature-dependent Sellmeier equations. Changing the temperature of the crystal from 20°C to 130°C enables signal tuning from 1284 nm to 1270 nm, while the idler can be tuned from 6200 nm to 6551 nm. Although these calculations are based on Eq. (1) and Eq. (2), investigations for accurate dispersion properties of this new nonlinear material resulted in variants of sellmeier equations.[14]

3. EXPERIMENTAL DESIGN AND SETUP

The schematic of the experimental setup is shown in Fig. 2. The high-energy MOPA pump laser system is seeded by a 450 MHz passively mode-locked oscillator providing 5.4 ps micro-pulses. An acousto-optic modulator selects a train of micro-pulses with 1-µs duration (macro-pulse) at a repetition rate of 20 Hz, and successive amplifier stages increase the macro-pulse energy up to 50 mJ, corresponding to an average power of 1 W. This represents a single micro-pulse energy of 0.1 mJ with a measured pulse width of 8.6 ps at a central wavelength of 1064 nm.[15]

An important parameter governing the OPO conversion efficiency is the pump spectral acceptance bandwidth for parametric generation in CSP under type-I (*e*→*oo*) NCPM. The pump laser operates at a central wavelength of 1064.1

nm and has an FWHM spectral bandwidth of 0.16 nm. This is shown in Fig. 4, relative to the parametric gain bandwidth of 0.33 nm for 12.1-mm-long CSP crystal used in our experiment, estimated using the relevant Sellmeier equations.[5,12] This confirms that the pump bandwidth is not a factor limiting the efficiency in our CSP OPO. The output beam from the pump laser has a diameter of 2 mm and a beam quality factor of M^2~1.1. The CSP crystal is a 4-mm-wide (along the c-axis) and 5-mm-thick sample, grown from stoichiometric melt by the horizontal gradient freeze technique.[5]

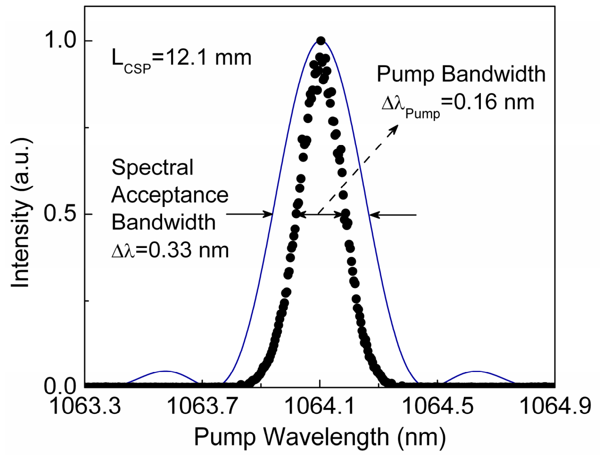

Figure. 4. Pump laser spectrum relative to parametric gain bandwidth for 12.1-mm-long CSP crystal.

It is cut at θ=90°, Φ=45° for type-I (*e→oo*) NCPM and housed in an oven with temperature stability of ±0.1°C. Both crystal faces are antireflection coated with a single layer sapphire coating, providing high transmission (T>98.7%) for the pump and signal over 1064-1300 nm and T>76% for the idler over 6000-6500 nm.

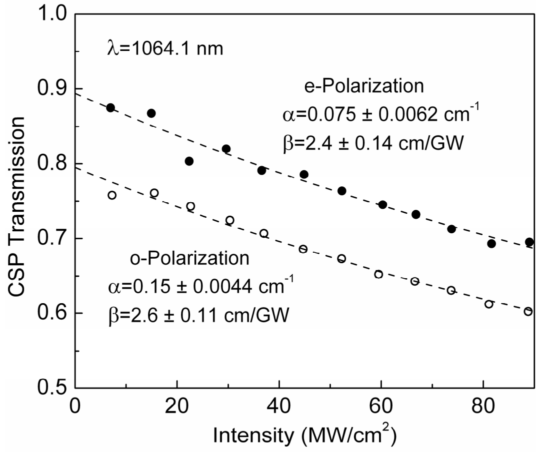

Figure. 5. CSP transmission as a function of pump intensity for *o*- and *e*-polarization at 1064.1 nm.

The residual loss at 1064.1 nm is estimated from the measured transmission of the crystal. As depicted in Fig. 5, we observed a drop in the transmission of CSP crystal with increasing pump intensity, showing a nonlinear behavior. Using a simple two-photon absorption model, we fitted the measured data for linear (α) and two photon (β) absorption coefficients, resulting in a value of α=0.075 cm^{-1}, β=2.4 cm/GW for extraordinary (*e*) and α=0.15 cm^{-1}, β=2.6 cm/GW for ordinary (*o*) polarizations. These values indicate the improved quality of this crystal as compared to the earlier samples.[6-11] Further, using this data, we have estimated the energy band-gap (E_g) of the material, resulting in the values of 2.08 eV (*e*) and 2.06 eV (*o*), confirming the large band-gap of CSP.[16] The OPO is configured as a singly-resonant oscillator in a compact linear standing-wave cavity comprising two curved mirrors, M_1 and M_2, with radius of curvature r=3 m (ZnSe substrate). Both mirrors are highly reflecting (R>99%) for signal over 1200-1400 nm, and highly transmitting at 1064 nm (T>97%) and for the idler over 5500-7500 nm (T>98%). The pump beam has a waist radius of w_0~1.5 mm after the input mirror (M_1) to avoid any damage, while using the maximum aperture of the crystal. A dichroic mirror, M_3, highly reflecting (R>99%) at 1064 nm and highly transmitting (T>95%) for the idler, separates the generated idler from the undepleted pump. In the measurements of energy and efficiency, all the data were corrected for transmission and reflection coating losses. The total round-trip optical length of the cavity, including the CSP crystal, is 666 mm, corresponding to a 450 MHz repetition rate, ensuring the synchronization with the pump laser.

4. RESULTS AND DISCUSSION

4.1 Tuning

We characterized the OPO with regard to output pulse energy and tunability by varying the crystal temperature at a constant pump energy of 30 mJ. Figure 6 shows the generated idler wavelengths as a function of temperature together with the theoretically estimated wavelengths calculated from Eq. (1) and Eq. (2). The idler wavelength varies at a rate of 3 nm/°C. The discrepancy between the calculated and experimental data points could be due to Eq. (2), indicating that the temperature dependent Sellmeier equations have to be further refined. Figure 7 shows the extracted idler energy as well as the transmission of CSP crystal across the tuning range. Using a 500 µm fused silica etalon, we extracted part of the intracavity signal to monitor the wavelength. The signal wavelength was recorded by using a low-resolution (10 nm) InGaAs spectrometer (Ocean optics, NIR Quest), and was further confirmed by single-pass second harmonic generation into the red in a 5 mm type-I (*oo*→*e*) BBO crystal. The idler wavelength was inferred from the second harmonic of the signal, which was measured using a high resolution (1 nm) CCD array spectrometer (Ocean optics, USB 4000). By changing the CSP crystal temperature from 30 °C to 180 °C, we could tune the idler wavelength from 6091 to 6577 nm, corresponding to a total tuning range of 486 nm. The generated idler energy varies from 1.3 mJ at 6091 nm to 1 mJ at

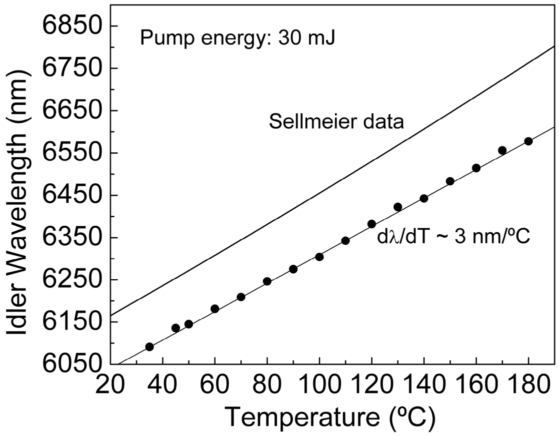

Figure. 6. Temperature tuning of CSP OPO.

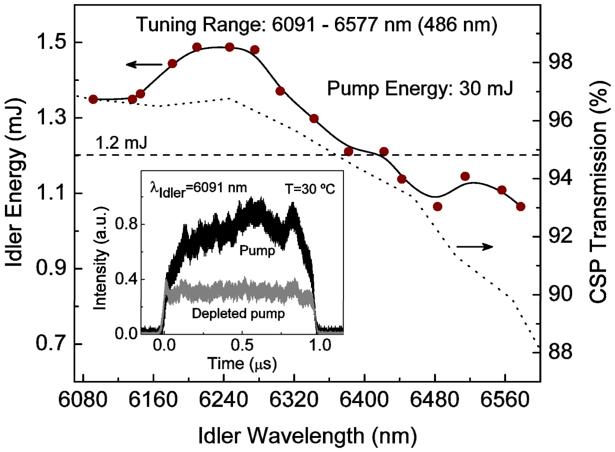

Figure. 7. Idler energy and CSP transmission across the tuning range. Inset: Macro-pulse envelope of input pump and depleted pump.

6577 nm, reaching a maximum of 1.5 mJ at 6275 nm, with >1.2 mJ over >68% of the tuning range. This represents a maximum idler energy conversion efficiency of 5% and a photon conversion efficiency of 29.5%. The drop in the idler energy towards the longer wavelengths is attributed to the water absorption peak near 6.4 µm and residual multi-phonon absorption in the CSP crystal, resulting in reduced transmission, as evident from Fig. 7. The corresponding pump depletion is recorded to be >42% over more than 64% of the tuning range with a maximum pump depletion of 51% at 6483 nm. Also shown in the inset of Fig. 7 is the macro-pulse envelope of the input and the depleted pump, measured using a fast photodiode, at 30 °C, corresponding to an idler wavelength of 6091 nm, clearly showing >50% depletion.

4.2 Energy scaling

We performed idler energy scaling measurements at different wavelengths across the OPO tuning range. The variation of the idler energy and pump depletion as a function of the pump energy obtained at a temperature of 30 °C, corresponding to an idler wavelength of 6091 nm, is shown in Fig. 8. As evident from the plot, 1.35 mJ of idler is obtained for a pump energy of 30 mJ at a slope efficiency, η=4.6%, implying a peak idler energy efficiency of 4.5%, representing a photon conversion efficiency of 25.7%. The threshold pump energy is measured to be 0.7 mJ, corresponding to single micro-pulse energy of 1.5 µJ, and strong pump depletion reaching >50% is achieved above an input pump energy of 5 mJ. The peak efficiency in this experiment was limited by the low optical damage threshold of the single layer sapphire coating on the CSP crystal, observed beyond 30 mJ of pump energy. Increasing the pump energy to 31.5 mJ, representing a peak intensity of 100 MW/cm^2, resulted in surface damage on the input face of the crystal, while no damage was observed on the exit face, indicating that the damage is due to the pump beam. Although the damage did not prevent OPO operation, a substantial drop of 37% in the idler energy was recorded. We also characterized the OPO near 6400 nm, a technologically important wavelength for surgical applications[1], as shown in the inset of Fig. 8. At 130 °C, corresponding to an idler wavelength of 6422 nm, an idler energy up to 1.1 mJ is generated for a pump energy of 30 mJ at a slope efficiency of η=3.8%, with an increased threshold pump energy of 1.6 mJ due to water absorption and reduced crystal transmission.

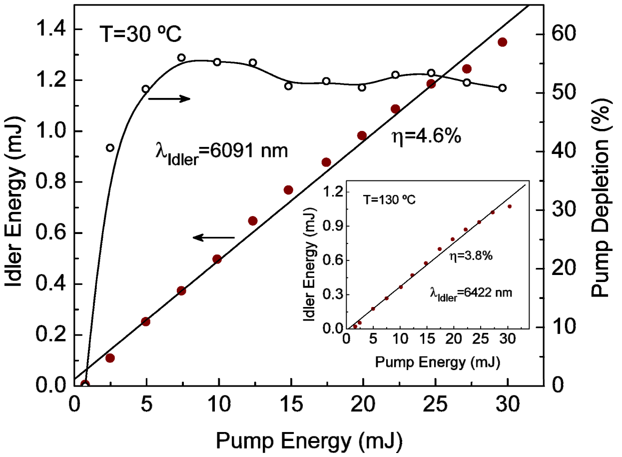

Figure. 8. Idler energy scaling and pump depletion in CSP OPO as a function of input pump energy at 6091 nm. Inset: Idler energy scaling at 6422 nm.

4.3 Spatial and temporal characteristics

We determined the duration of the signal pulses extracted from the OPO using a home-made autocorrelation setup. Figure 9(a) shows the measured autocorrelation profile at 1289 nm, where the amount of signal energy extracted from the cavity was >1.3 mJ for an incident pump energy of 30 mJ. The FWHM of the trace is 16.3 ps, resulting in the signal pulse duration of 10.6 ps, assuming a sech^2 pulse shape. This value of pulse duration was confirmed by repeating the measurement several times. Similar pulse duration is expected across the tuning range. Also shown in the inset of Fig. 9(a) is the signal beam spatial profile at 1289 nm, measured using a pyroelectric camera (Spiricon, Pyrocam-III). The corresponding idler beam profile at 6091 nm, recorded at full output energy, is shown in Fig. 9(b). Both signal and idler beam profiles confirm good beam quality with Gaussian profiles, which is important for surgical applications.

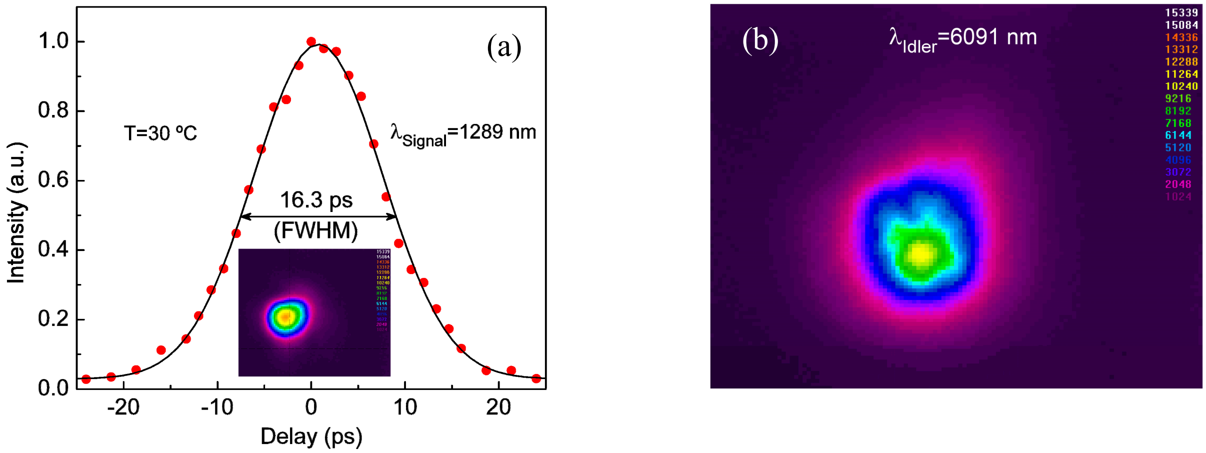

Figure. 9. (a) Typical autocorrelation trace of the CSP OPO output signal pulse at 1289 nm, resulting in a duration of 10.6 ps (×1.54, assuming sech^2 pulse shape). Inset: Signal beam profile at 1289 nm, (b) Corresponding idler beam profile at 6091 nm.

5. CONCLUSIONS

In conclusion, we have demonstrated a compact, high-energy, high-repetition-rate picosecond OPO for the mid-IR based on the new nonlinear material CSP, pumped at 1064.1 nm. The OPO delivers as much as 1.5 mJ of idler energy at 6275 nm and is tunable over 486 nm across the technologically important wavelength range of 6091-6577 nm for surgical applications, with >1.2 mJ over >68% of the tuning range. The compact design, high energy, suitable pulse structure, and the potential for energy scaling using larger aperture crystals, make the OPO a promising source for practical surgical applications in the mid-IR.

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