

# Narrow-bandwidth, mid-infrared, seeded optical parametric generation in 90° phase-matched CdSiP<sub>2</sub> crystal pumped by diffraction limited 500 ps pulses at 1064 nm

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Low-threshold, efficient optical parametric generation at ~6100 nm is demonstrated using CdSiP<sub>2</sub> nonlinear crystal at 1 to 10 kHz repetition rates with relatively long 500 ps pump pulses at 1064 nm. Maximum single pulse energy of 8.7 μJ and average power of 79 mW are achieved for the idler. Seeding at the signal wavelength is employed using a distributed feedback laser diode, which enables approximately tenfold narrowing of the idler bandwidth down to less than 1 nm. © 2012 Optical Society of America

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Starting from spontaneous parametric fluorescence, the threshold of conventional traveling-wave type optical parametric generators (OPGs) can only be reached with the high peak power available from ultrashort (femtosecond or picosecond) pump sources for which a cavity [optical parametric oscillator (OPO)] would be impractical (10 ps corresponds to only 3 mm in free space). Longer pulses, however, are interesting for narrow bandwidths, and one of the OPG advantages is that, in comparison to OPO, seeding is much easier to apply for narrowband single-frequency operation [1]. It is clear that high-gain second-order nonlinear materials are attractive for such temporal regimes. The new chalcopyrite type nonlinear crystal CdSiP<sub>2</sub> (CSP) possesses extremely high (84.5 pm/V) second-order nonlinear coefficient and can be pumped at 1064 nm in a noncritical configuration without two-photon absorption. Thus, it is of special interest for direct access to the mid-IR spectral range up to ~6.5 μm [2]. Highly efficient unseeded OPG with CSP has already been demonstrated using 8.7 ps pump pulses at 1064 nm from a commercial Nd:YVO<sub>4</sub> mode-locked laser/amplifier system operating at 100 kHz [3], achieving bandwidths of 8.5 nm for the signal at 1282 nm and 122 nm for the idler at 6204 nm. On the other hand, pumping with 8 ns pulses at 1064 nm, we observed extremely low OPG threshold in CSP (<2 mJ/cm<sup>2</sup> axial fluence), however, residual idler reflections from the antireflection (AR) crystal coatings played the role of a low-finesse OPO resonating the idler, which complicated the interpretation of the results [4]. In the present work, we apply 500 ps pump pulses for which OPO feedback is impossible and confirm the extremely low OPG threshold of CSP. Moreover, employing suitable diode laser seeding we demonstrate narrowband noncritical OPG operation at 1 to 10 kHz repetition rates. Thus the longer pump pulses in comparison to [3] are associated not only with simplified pump system design based on Q-switching, but also ensure ultimately much narrower spectral bandwidths in the mid-IR.

The CSP crystal available for the present experiment was 21.4 mm long with an aperture of 4.1 mm (along *c*) × 6.1 mm. It was cut for noncritical (90°) Type I (oo-e) interaction, as shown in Fig. 1(a). Both faces were AR coated with a single layer of Al<sub>2</sub>O<sub>3</sub> (TwinStar), which in fact was optimized only for the pump and signal wavelengths (minimum reflection of ~1% at 1150 nm), hoping in this way to achieve a higher surface damage threshold. The actual reflectivity measured at the three wavelengths was 1.3% (1064 nm), 2% (1288 nm), and 20% (6125 nm) per surface.

The experimental setup is shown in Fig. 1(b). The pump source was a diode-pumped laser system consisting of a passively Q-switched Nd:YAG oscillator with pulsed diode pumping and a double-pass side-pumped

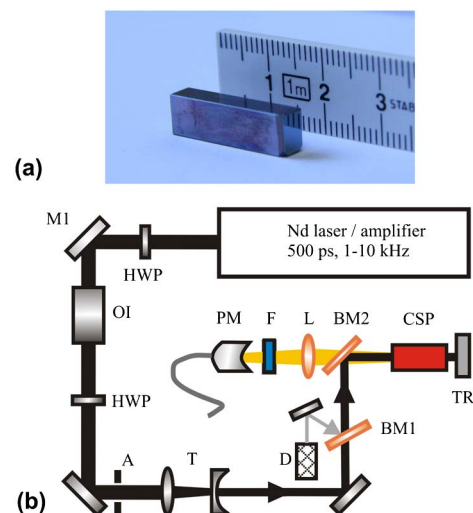


Fig. 1. (Color online) (a) Photograph of the AR-coated CSP element and (b) OPG experimental setup: M1, polarization dependent reflection mirror; HWP, half-wave plate; OI, optical isolator; T, telescope; A, variable size aperture; D, DFB seed diode; BM1 and BM2, dichroic bending mirrors; TR, total reflector; L, 10 cm BaF<sub>2</sub> lens; F, 5 μm cut-on filter; PM, power meter.

Nd:YVO<sub>4</sub> amplifier described in detail elsewhere [5]. The maximum available pump energy was about 470  $\mu\text{J}$ . The pump beam from the amplifier was downcollimated by a spherical lens telescope to a slightly elliptical shape with approximately Gaussian spatial distribution and diameters of  $2w \approx 1.38$  mm and  $2w \approx 1.15$  mm in the horizontal (parallel to the CSP  $c$ -axis) and vertical directions, respectively. Dichroic bending mirrors (BM1 and BM2) on ZnSe substrates were used to recombine the seed and pump beams and to separate the idler beam in a double-pass OPG arrangement with an Ag total reflector for the three waves. BM1 transmits 95% of the pump radiation and BM2 reflects about 98% at 45°. With 84% transmission of the telescope, this resulted in maximum pump pulse energy at 1064 nm of  $\sim 370$   $\mu\text{J}$  incident on the CSP crystal. The pump pulse duration was  $\sim 500$  ps. The seed diode was a fiber-coupled wavelength stabilized distributed-feedback (DFB) laser diode (QPHOTONICS) with a maximum power of 2 mW near 1290 nm. Since BM2 only partially reflects the signal ( $\sim 45\%$ , polarizing the seed in the desired direction), and the diode was not operated at maximum current, about 0.5 mW of seed power was incident on the CSP crystal with proper polarization. With the built-in Peltier cooler, it was possible to slightly decrease the wavelength, matching the CSP parametric fluorescence. The reported OPG energies (signal and idler) were corrected for the transmission of the bending mirror BM2. The signal power was measured by substituting the filter  $F$  in Fig. 1 by a calcite polarizer that blocks the pump and absorbs the idler.

While in single-pass OPG operation (total reflector removed), the threshold (defined at a detection limit of  $\sim 20$   $\mu\text{W}$  for the idler average power) for seeded operation was more than two times lower and the output energies drastically increased in the presence of seeding [Fig. 2(a)]. In double-pass operation, the input–output characteristics were almost independent of the seed [Fig. 2(b)]. The threshold for unseeded double-pass OPG operation was 43  $\mu\text{J}$ , i.e., 6.9  $\text{mJ}/\text{cm}^2$  of peak on-axis pump fluence or 13.8  $\text{MW}/\text{cm}^2$  of on-axis intensity [Fig. 2(b)]. In terms of fluence, this is even lower than the record value of 8.2  $\text{mJ}/\text{cm}^2$  achieved with 2  $\mu\text{J}$ , 10 ns pump pulses at 3.1  $\mu\text{m}$  when pumping a noncritically cut 24 mm long type-II ZnGeP<sub>2</sub> (ZGP) crystal in an OPO with a similar double-pump pass arrangement [6]. Note that in our case (where CSP and ZGP have very similar effective nonlinearity), OPO feedback is impossible because the index of refraction of CSP ( $\sim 3.1$ ) results in a cavity round trip time of 442 ps if the residual reflectivity of the AR coatings is considered, comparable to the pump pulse FWHM. When the seed laser diode was switched on, the threshold of the double-pass arrangement decreased to 31.5  $\mu\text{J}$  of incident pump energy (5  $\text{mJ}/\text{cm}^2$  on-axis fluence and 10  $\text{MW}/\text{cm}^2$  on-axis intensity thresholds), as shown in Fig. 2(b).

The maximum conversion efficiency in the double-pass arrangement reached  $\sim 20\%$  [Fig. 3(b)]. This dependence was saturated (to the same level also in the absence of seeding), and the difference between measured pump depletion and estimated total conversion efficiency was marginal if compared to the single-pass case [Fig. 3(a)]. The maximum idler energy at 1 kHz in the double-pass arrangement, achieved at a pump energy of

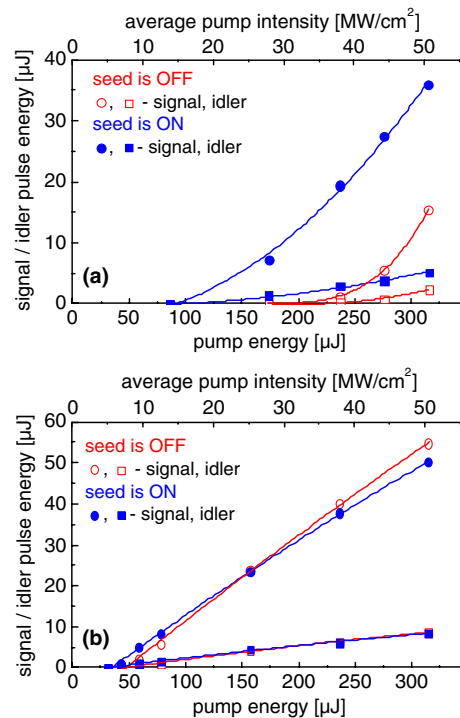


Fig. 2. (Color online) Input–output characteristics of the CSP OPG at 1 kHz in (a) single-pass and (b) double-pass arrangement. Deviations of the ratio signal to idler energy from the value determined by energy conservation can be attributed to the idler residual reflection losses at the CSP surfaces.

316  $\mu\text{J}$ , amounted to 8.7  $\mu\text{J}$  and 8.5  $\mu\text{J}$  in the unseeded and seeded regime, respectively [Fig. 2(b)]. This pump level corresponds to on-axis values of  $>100$   $\text{MW}/\text{cm}^2$  and  $>50$   $\text{mJ}/\text{cm}^2$  for the intensity and fluence, respectively. Thus, no damage was observed at pump intensities at

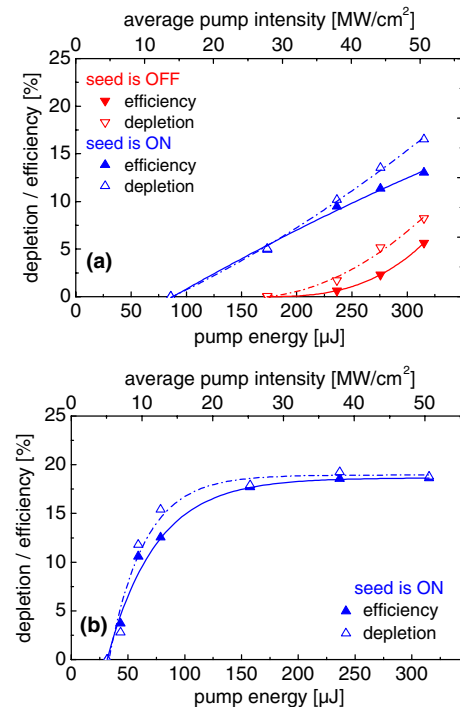


Fig. 3. (Color online) Total conversion efficiency (signal + idler) and pump depletion at 1 kHz in (a) single-pass and (b) double-pass arrangement.

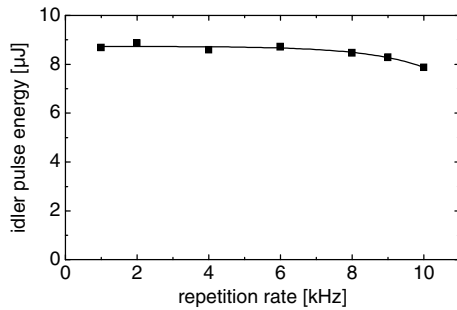


Fig. 4. Idler pulse output energy versus repetition rate for incident pump energy of 300  $\mu\text{J}$  (seeded).

least four times higher than in [2], which can be attributed to the shorter pulse durations in the present experiments; note that in [2], surface damage occurred at an on-axis fluence of about 350  $\text{mJ}/\text{cm}^2$ .

The output energy changed only slightly when increasing the repetition rate, decreasing only when approaching 10 kHz as can be seen from Fig. 4, where the pump beam spatial profile exhibits some changes. Nevertheless, the maximum average power at this repetition rate reached 79 mW for the idler.

Pump and signal spectra were recorded with an AQ6317B (Ando Electric) spectrum analyzer with maximum possible resolution of  $<15$  pm. The single-pump pulse is single frequency, and therefore should be Fourier limited [5], but due to thermomechanical noise, the pulse frequency is not fixed and can drift within the bandwidth measured with the spectrum analyzer [Fig. 5(b)], which is still less than the 60 pm oscillator free-spectral range. However, keeping in mind the specified DFB laser linewidth of  $<10$  pm and the close values for the pump and seed bandwidths in Fig. 5(b), it can be assumed also that we measure in these cases the actual spectral response of the spectrum analyzer, which should be around 30 pm. At maximum output level, the bandwidth of the OPG signal spectrum decreased from 400 pm (when the seed is off) to 52 pm (when the seed is on) in the double-pass scheme (Fig. 5); in the single-pass arrangement, this bandwidth reduction was from  $\sim 300$  pm to 35 pm, very close to the actual spectral resolution, which means that both narrowing factors represent only lower limits. The effect of the seeding on the narrowing of the signal output spectrum is shown in Fig. 5(a) for the double-pass scheme. Since in the double-pass scheme the actual pump bandwidth is much narrower than the measured signal bandwidth, one can conclude that the idler bandwidth reproduces the signal bandwidth in terms of wavenumbers. Deconvolving the latter with a spectral function of 30 pm leads to an estimation of 42 pm for the signal, which translates into 0.94 nm for the idler generated at 6102 nm. The signal pulse duration in the double-pass seeded regime measured by a fast photodiode was  $\sim 300$  ps. Temporal walk-off effects can be ignored on the present time scale, or, in other words, there is an excess of spectral acceptance, which is why the pulse duration of the idler should be very close to that of the signal pulse. Under these assumptions, the Fourier product, both for signal and idler, has an upper limit of  $\sim 2.3$ . Note that this product was 9.3 in [3], where no bandwidth control was applied but the spectral extent was rather deter-

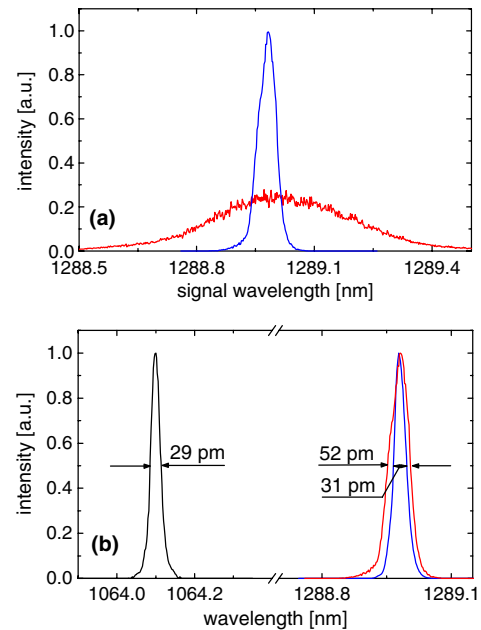


Fig. 5. (Color online) (a) Unseeded (lower red curve) and seeded OPG signal spectrum (tall blue curve) in the case of double-pass operation, and (b) pump (black curve), seed diode (blue curve), and output signal (red curve) spectra.

mined by the parametric gain bandwidth (or spectral acceptance), i.e., primarily by the crystal length. Although, as already explained, the  $\sim 2.3$  Fourier product in the present work is possibly a result of time averaging, this is the value relevant for practical applications of the system.

In conclusion, pumping a relatively long noncritically phase-matched CSP crystal with 500 ps pulses at 1064 nm, OPG idler energies near 6100 nm as high as 8.7  $\mu\text{J}$  and maximum average power of 79 mW were achieved at variable repetition rate of 1 to 10 kHz. Seeding at the signal wavelength with a DFB laser diode enabled  $\sim 10$  fold narrowing of the idler bandwidth down to  $<1$  nm, leading to a time-averaged Fourier product of  $\sim 2.3$ , which, in terms of spectral resolution, is an improvement of  $\sim 130$  times as compared to previous results obtained with sub-10 ps pulses. Wavelength tuning of this system would require temperature tuning of both the CSP crystal and the seed diode.

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