Noncritical singly resonant synchronously pumped OPO for generation of picosecond pulses in the mid-infrared near 6.4 μm

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The recently developed chalcopyrite $CdSiP_2$ is employed in a picosecond, 90°-phase-matched, synchronously pumped, optical parametric oscillator pumped at 1064 nm to produce steady-state idler pulses near 6.4 μm with an energy as high as 2.8 μJ at 100 MHz, in a train of 2-μs-long macropulses following at a repetition rate of 25 Hz. Without an intracavity etalon, the 12.6-ps-long micropulses have a spectral width of 240 GHz.

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Synchronously pumped optical parametric oscillators (SPOPOs) represent a potentially efficient source of high-repetition-rate (~ 100 MHz) ultrashort pulses at wavelengths not available from conventional mode-locked lasers. Difference-frequency generation (DFG) can also be used to this aim but yields very low quantum efficiency and requires two synchronized wavelengths. Depending on the pump source, e.g., mode-locked Ti:sapphire and Nd lasers or their second harmonics, SPOPOs normally generate 100 fs–100 ps-long pulses. Being downconversion devices, their coverage in the mid-IR is mostly limited to 4–5 μm, the absorption edge of oxide-based nonlinear crystals. Normally, the signal wavelength is only resonated, and although in some special cases (high parametric gain in the exit layer of periodically poled $LiNbO_3$, PPLN) the generated idler extended up to 7.25 μm [1], the single-pulse energy at such wavelengths is extremely low (~ 0.3 pJ).

The use of chalcogenide crystals, transparent in the mid-IR, has been reported in only a few cases. Preserving the high repetition rate was possible only by cascaded operation of two SPOPOs. Thus, a noncritically phase-matched CdSe-based SPOPO was pumped and tuned by the signal wave of a PPLN-based SPOPO at 120 MHz and generated picosecond idler pulses, but the wavelength range was limited to 9.1–9.7 μm and the maximum single pulse energy was 90 pJ [2]. Another SPOPO based on $AgGaSe_2$ was pumped at 1.55 μm by the signal wave of an 82 MHz femtosecond $CsTiOPO_4$-based SPOPO and generated 0.4–0.5-ps-long pulses up to 7.9 μm, with single-pulse energy of 270 pJ at 5.25 μm [3].

In general, the cavity-length stabilization and synchronization of three short-pulse devices (mode-locked pump laser and two SPOPOs) is a serious problem that is detrimental for long-term stability and potential applications. Thus, for attainment of devices with improved practicability, compact design, and utility, it is of great interest to develop mid-IR SPOPOs based on chalcogenide materials that can be directly pumped by conventional mode-locked lasers operating in the near-IR, especially near 1 μm. However, nonlinear crystals like the commercially available $AgGaS_2$ (AGS), which can be used at 1064 nm without the onset of two-photon absorption (TPA), have poor thermomechanical characteristics like thermal conductivity, anisotropy of the thermal expansion, and damage threshold. Three concepts to reduce the load to the nonlinear material in terms of average power have been implemented so far: a 32-fold mechanical chopper was used at 40 Hz to reduce the average power of the 20 W, 100 ps Nd-YAG pump laser operating at 76 MHz [4]; a similar mode-locked laser was additionally Q switched at 2 kHz to produce an average power of up to 8.5 W at a repetition rate of 2 kHz for the nonstationary macropulses consisting of about 20 micropulses of 145 ps duration [5]; and a pulsed Nd-YAG oscillator-amplifier system, generating about 100 micropulses of 12.5 ps duration with an average power of up to 1.2 W, at 25 Hz for the macropulses, was employed in [6]. Note that these approaches lead to substantial increase of the single-pulse energy that reached the nanojoule [4] and microjoule [5,6] level in the 4–5.5 μm wavelength range for the idler.

The recently discovered cadmium silicon phosphide, $CdSiP_2$ (CSP) [7], is a negative uniaxial II–IV–V_2 chalcopyrite compound (space group 42 m) that allows 1064 nm pumping without TPA and possesses a useful transparency up to 6.5 μm, limited by intrinsic multiphonon peaks. As shown in [8], it outperforms all other mid-IR nonlinear materials that can be pumped near 1 μm in almost every aspect, with the main problem (yet to be solved) being the residual absorption close to the bandgap, which is not intrinsic. In addition, it is the only material that, without being a solid solution, still allows noncritical phase matching when pumped near 1 μm, with a maximum effective nonlinearity of $d_{\mathrm{eff}}=d_{36}=84.5$ pm/V [8,9]. Here, we demonstrate the first SPOPO based on 90°-cut CSP, pumped at 1064 nm.

The sample used in the present study (Fig. 1) was grown by directional solidification in a modified, high-temperature transparent furnace using the horizontal gradient freeze technique. It was cut at $\theta=90°$, $\varphi=45°$ and had a length of 9.5 mm. Its aperture was 6 mm (along the c axis) × 6.75 mm. The residual losses measured for the relevant polarizations (e for the pump and o for the signal and idler) are 0.185 cm^{-1} at 1064 nm, 0.114 cm^{-1} at 1.3 μm, and 0.014 cm^{-1} at 6.4 μm. Both faces were antireflection-coated for the three wavelengths (pump, signal, and idler) and the eight-layer coating (TwinStar) resulted in averaged reflectivity per surface of ~0.35% at 1064 nm, ~0.4% at 1275 nm, and ~0.8% at 6400 nm.

The pump source was an upgraded version of the one described in [6]; more details will be provided in a separate publication. It basically consisted of a flashlamp-pumped Nd:YAG oscillator operating at 25 Hz, mode locked at 100 MHz by a combination of acousto-optical modulator and a nonlinear mirror stabilized with passive negative feedback (two-photon absorber), followed by an external acousto-optical modulator to cut the initial (nonstationary) part of the pulse train and a three-pass amplifier stage, based again on Nd:YAG, to boost the energy. The system is capable of generating up to ~2.5 W of average power. This corresponds to energy of 100 mJ for a 2-μs-long macropulse consisting of 200 micropulses with a separation of 10 ns or energy of 0.5 mJ per micropulse for duration in the 15–20 ps range. The SPOPO cavity consists of two RC=−3 m curved mirrors for the signal only, at a separation of 1.5 m, giving a Gaussian mode diameter of 3.2 mm in the middle, where the nonlinear crystal was placed. The beams were separated by slightly noncollinear geometry, where the external angle between the pump and the cavity axis was about 2°. The pump beam diameter was ~2 mm.

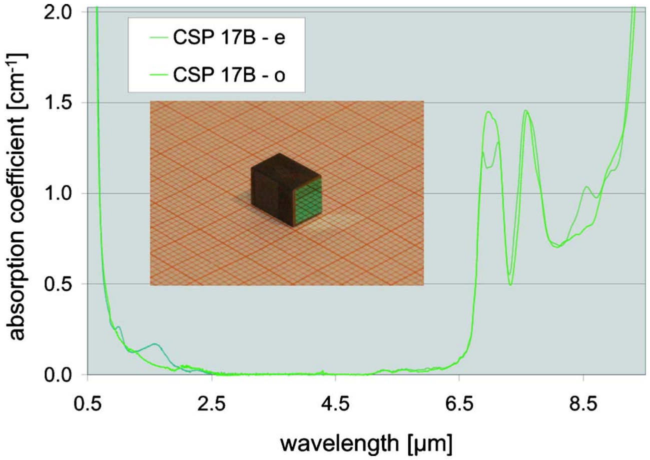

Fig. 1. (Color online) Polarized transmission of the CSP 17B sample used, measured prior to coating. Inset, photograph of the AR-coated sample.

We estimated the idler energy both from the pump depletion and using a calibrated pyroelectric detector. The threshold corresponded to an average pump power of 15 mW or a single (micro)pulse energy of 3 μJ. The depletion together with the simultaneously measured transmission of the crystal is shown in Fig. 2(a), while Fig. 2(b) shows the input–output characteristics.

The incident pump power was limited to slightly above 300 mW, because strong pump depletion, reaching typically >40%, was observed starting from about 100 mW of pump power. The actual crystal transmission at the pump wavelength, when the OPO cavity was blocked, was lower than expected, but this was confirmed in additional extracavity measurements, using also a cw source with comparable beam size, and can be explained by inhomogeneous crystal quality (defects inside). However, no signs of TPA are seen in Fig. 2(a). The directly measured idler power was in good agreement with estimations from the pump depletion, Fig. 2(b). The maximum idler power of 14 mW corresponds to a single (micro)pulse energy of 2.8 μJ.

Only normal incidence was studied, but the slightly noncollinear interaction resulted in ~200 nm longer idler wavelengths. From the measured signal wavelength of 1276.55 nm (Fig. 3), one arrives at an idler wavelength of 6397.5 nm. This was confirmed by calculations using the refined Sellmeier equations [9], which predicted the same idler wavelength for an internal angle of 0.67° between the signal and the pump waves. The signal bandwidth (Fig. 3) corresponds to 8 cm^{-1} or 240 GHz, which is comparable,

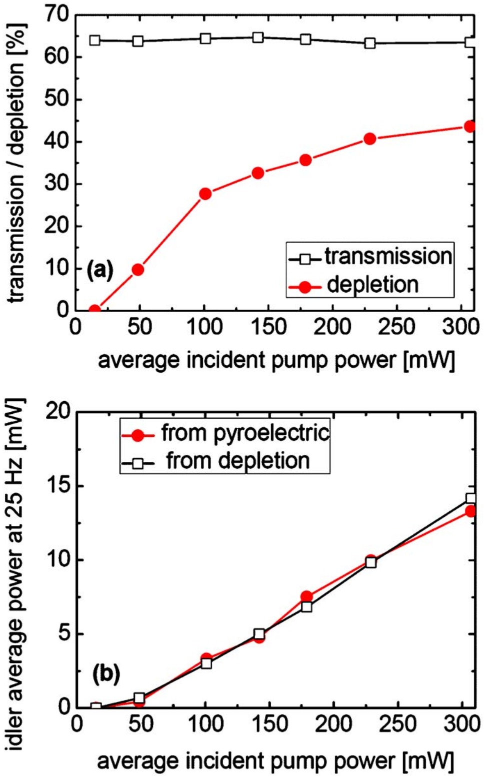

Fig. 2. (Color online) (a) Pump depletion and crystal transmission at 1064 nm versus pump power and (b) idler power versus pump power.

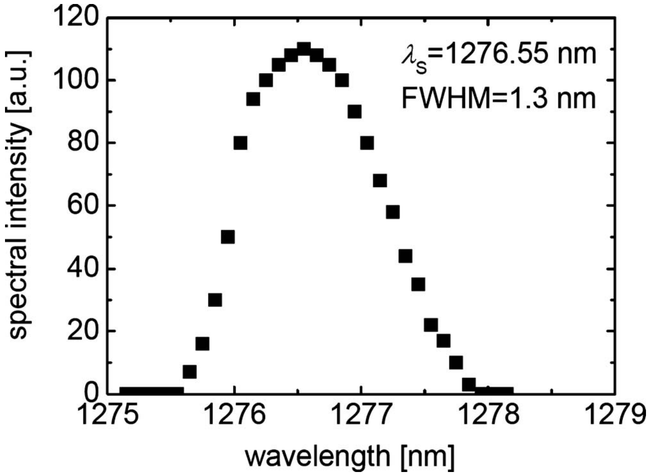

Fig. 3. Signal spectrum measured from a cavity leakage at a pump level of 200 mW, with 0.4 nm spectral resolution.

though slightly larger than the spectral acceptance of CSP for DFG.

We measured the duration of the idler pulses using noncollinear second-harmonic generation in a 2-mm-thick $HgGa_2S_4$ crystal cut at $\varphi=45°$, $\theta=40°$ for Type I interaction. A ZnSe plate served as a beam splitter, and the second harmonic was detected by a PbS resistor connected to a lock-in amplifier. Figure 4 shows the result of averaging four traces.

The Lorentzian fit is better than the Gaussian one and gives a FWHM ~25 ps for the trace, which means a pulse FWHM of 12.6 ps. Assuming the same spectral bandwidth for the idler, one arrives at a time–bandwidth product of 3, more than 1 order of magnitude above the Fourier limit.

Finally, by using a hair dryer, we confirmed that slight heating of the crystal results in shorter (about 2 nm) signal wavelength, which translates into an idler wavelength in the 6450 nm range, important for medical (surgical) applications [10]. This is an indication of the possibility for practical tuning by temperature variation in the noncritical configuration, which potentially can cover the transparency range up to the long-wave limit.

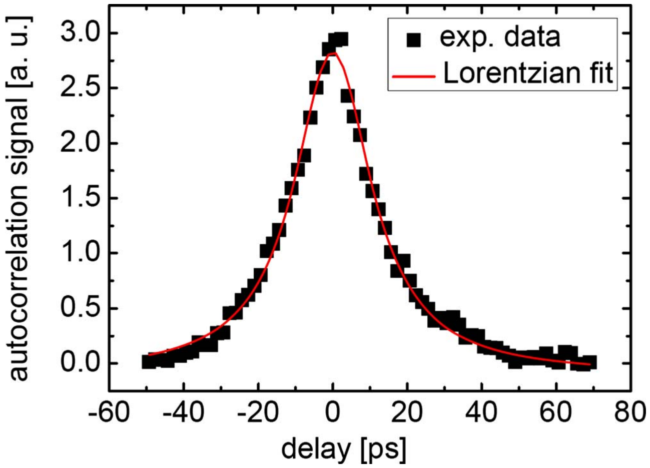

Fig. 4. (Color online) Experimental data (symbols) and Lorentzian fit (curve) of a background-free autocorrelation trace for the idler pulses.

When CSP was replaced by AGS ($10\times10\times10$ mm^3) cut for Type I phase matching, the parametric oscillation threshold increased to ~100 mW, i.e., about 1 order of magnitude. Moreover, the use of AGS in this SPOPO was accompanied by gradual surface blackening. Surface lifetime could be kept above ~400 h only if the pump average power did not exceed ~200 mW.

In conclusion, we demonstrated for the first time to our knowledge SPOPO operation of CSP pumped at 1064 nm. Future work will be focused on reduction of the residual loss, which alone is expected to lead to two-fold improvement of the output powers, temperature tuning maintaining the noncritical configuration, and power scaling using crystals of larger aperture.

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References

1. M. A. Watson, M. V. O'Connor, P. S. Lloyd, D. P. Sheperd, D. C. Hanna, C. B. E. Gawith, L. Ming, P. G. R. Smith, and O. Balachninaite, Opt. Lett. **27**, 2106 (2002).
2. M. A. Watson, M. V. O'Connor, D. P. Shepherd, and D. C. Hanna, Opt. Lett. **28**, 1957 (2003).
3. S. Marzenell, R. Beigang, and R. Wallenstein, Appl. Phys. B **69**, 423 (1999).
4. E. C. Cheung, K. Koch, and G. T. Moore, Opt. Lett. **19**, 631 (1994).
5. K. J. McEwan, Opt. Lett. **23**, 667 (1998).
6. A. A. Mani, L. Dreesen, Ph. Hollander, C. Humbert, Y. Caudano, P. A. Thiry, and A. Peremans, Appl. Phys. Lett. **79**, 1945 (2001).
7. P. G. Schunemann, K. T. Zawilski, T. M. Pollak, D. E. Zelmon, N. C. Fernelius, and F. K. Hopkins, in *Advanced Solid-State Photonics* (Optical Society of America, 2008) postdeadline paper MG6.
8. V. Petrov, F. Noack, I. Tunchev, P. Schunemann, and K. Zawilski, Proc. SPIE **7197**, 7197-21 (2009).
9. P. G. Schunemann, K. T. Zawilski, T. M. Pollak, V. Petrov, and D. E. Zelmon, in *Advanced Solid-State Photonics* (Optical Society of America, 2009), paper TuC6.
10. G. S. Edwards, R. H. Austin, F. E. Carroll, M. L. Copeland, M. E. Couprie, W. E. Gabella, R. F. Haglund, B. A. Hooper, M. S. Hutson, E. D. Jansen, K. M. Joos, D. P. Kiehart, I. Lindau, J. Miao, H. S. Pratisto, J. H. Shen, Y. Tokutake, A. F. G. van der Meer, and A. Xie, Rev. Sci. Instrum. **74**, 3207 (2003).