Subnanosecond, 1 kHz, temperature-tuned, noncritical mid-infrared optical parametric oscillator based on $CdSiP_2$ crystal pumped at 1064 nm

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Operation of an optical parametric oscillator based on $CdSiP_2$ and pumped at 1064 nm is demonstrated at a repetition rate of 1 kHz. The maximum output idler energy of 24 μJ at 6.125 μm corresponds to an average power of 24 mW. Increasing the crystal temperature up to 150° in the noncritical (90°) configuration leads to idler wavelength tuning from 6.117 to 6.554 μm. Subnanosecond pulse durations are obtained for the signal and idler as a result of the 1 ns pulse duration of the pump, made possible by the rather short crystal and cavity lengths (~1 cm). © 2010 Optical Society of America

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The mid-IR spectral range can be continuously covered by nonlinear frequency down conversion using near-IR laser pump sources, but oxide crystals perform well only up to ~4 μm because multiphonon absorption sets on at longer wavelengths. For obtaining high average power and single-pulse energy, optical parametric oscillators (OPOs) are free of restrictions related to the spectral gain bandwidth or higher-order nonlinear effects, typical for ultrashort laser pulses, but even in that case the nonlinear crystals should possess a sufficiently large bandgap in order to avoid two-photon absorption (TPA) at the pump wavelength. This requirement is met by only few chalcogenide materials. The recently discovered cadmium silicon phosphide, $CdSiP_2$ (CSP) [1], is a negative uniaxial II–IV–V_2 chalcopyrite compound (space group $\bar{4}2m$) that enables 1064 nm pumping without TPA with a useful transparency up to 6.5 μm, limited by intrinsic multiphonon peaks. As shown in [2], it outperforms all other materials 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 with a maximum effective nonlinearity of $d_{\text{eff}}=d_{36}=84.5$ pm/V [3]. Recently, we demonstrated the first 90°-phase-matched singly resonant OPO based on CSP, pumped by 14 ns pulses at 1064 nm [4], which generated idler pulses at a fixed wavelength near 6.2 μm with an energy as high as 470 μJ, at a repetition rate of 10–20 Hz.

In this Letter we report three significant achievements with the CSP OPO: (i) the exceptionally high d_{eff} of CSP permits the use of rather short crystal/cavity lengths and consequently to pump with relatively short pump pulses, achieving, for the first time to our knowledge with any OPO, subnanosecond signal and idler pulse durations; (ii) the good thermomechanical properties enabled for the first time to our knowledge 1 kHz repetition rate operation with a non-oxide nonlinear material pumped at 1064 nm (the only previous attempt at 2 kHz [5] was in fact in the earliest report on such OPO based on proustite and resulted in immediate crystal damage; all further studies of chalcogenides until very recently were confined to 10 Hz or less); and (iii) we demonstrate that, in contrast to the related $ZnGeP_2$, temperature tuning is feasible for CSP providing an extension of the noncritical tuning range to the long; wavelength transmission limit, which covers the essential for medical applications spectral region near 6.45 μm [6].

The sample used in the present study [Fig. 1(a), inset] was cut at $\theta=90°$, $\varphi=45°$ and had a length of 9.5 mm. Its aperture was 6 mm (along the c axis) $\times$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 6.2–6.4 μm. Both faces were antireflection (AR)-coated for the three wavelengths (pump, signal, and idler) and the eight-layer coating (TwinStar) had average reflectivity per surface of ~0.35% at 1064 nm, ~0.4% at 1275 nm, and ~0.8% at 6.4 μm. A measurement prior to the OPO experiment gave a transmission of 77% at 1064 nm for the AR-coated sample.

The OPO cavity was the same as the one used in [4], Fig. 1(b). It consisted of two plane mirrors with a separation of 10 mm. The rear total reflector, TR, was an Ag mirror with a reflection coefficient of ~97%. The output coupler (OC) had a transmission of 20% at the signal and 75% at the idler wavelength; hence, the OPO can be considered as singly resonant with double-pass pumping. The CSP crystal was pumped through the OC, which transmitted 82% at 1064 nm. The beams were separated by the pump bending mir-

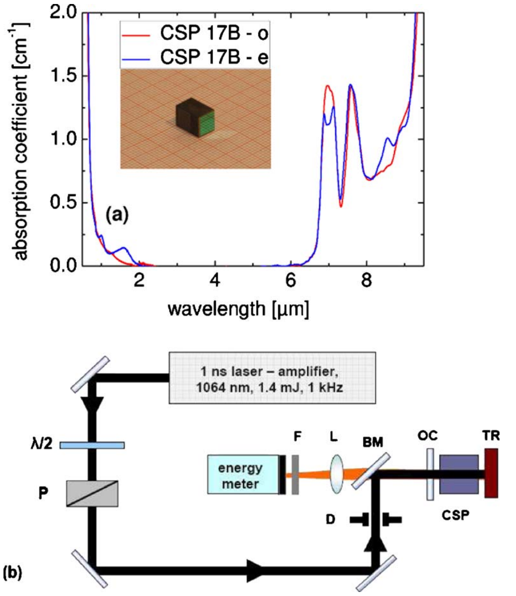

Fig. 1. (Color online) (a) Polarized transmission of the CSP 17B sample used, measured prior to coating, and a photograph of the AR-coated sample (inset). (b) Experimental setup of the CSP OPO: λ/2, half-wave plate; P, polarizer; F, 5 μm cut-on filter; L, 10 cm MgF_2 lens; D, diaphragm; BM, bending mirror; OC, output coupler; TR, total reflector.

ror (BM), which had high reflectivity for the pump (*s* polarization) and transmitted 82% and 84% (*p* polarization) at the signal and idler wavelengths, respectively. Both the plane-parallel OC and the BM were on ZnSe substrates with uncoated rear surfaces.

The pump source was a diode-pumped laser system consisting of an electro-optically *Q*-switched, 1 ns Nd:YVO_4 microlaser (ECR Model PML 1000), a cw-pumped Nd:YVO_4 regenerative amplifier, and a double-pass Nd:YAG post amplifier with pulsed pumping (both HighQLaser, Austria), optimized for a repetition rate of 1 kHz. The maximum available pump energy was about 1.4 mJ, of which 1.15 mJ were incident on the CSP crystal. A combination of a half-wave plate, λ/2, and a polarizer, *P*, served to adjust the pump energy. The pump beam had a Gaussian diameter of $2w \sim 3$ mm in the position of the OPO. Only the idler energy was measured behind the BM, the residual pump radiation and the signal were blocked by a 5 μm cut-on filter, F.

The OPO threshold was 120 μJ of pump energy (1.7 MW/cm^2 average pump intensity). The input-output characteristics are shown in Fig. 2(a). At 25°C, maximum idler energy of 24 μJ was achieved at 6.125 μm. This translates into an average power of 24 mW, an improvement of >2.5 times in comparison to our initial results at 10–20 Hz obtained with 14-ns-long pump pulses [4]. To the best of our knowledge, higher average powers (~28 mW) in the mid-IR have been reported with a 1064 nm pumped chalcogenide crystal in only one case, very recently with a $LiInSe_2$-based OPO operating at 100 Hz [7].

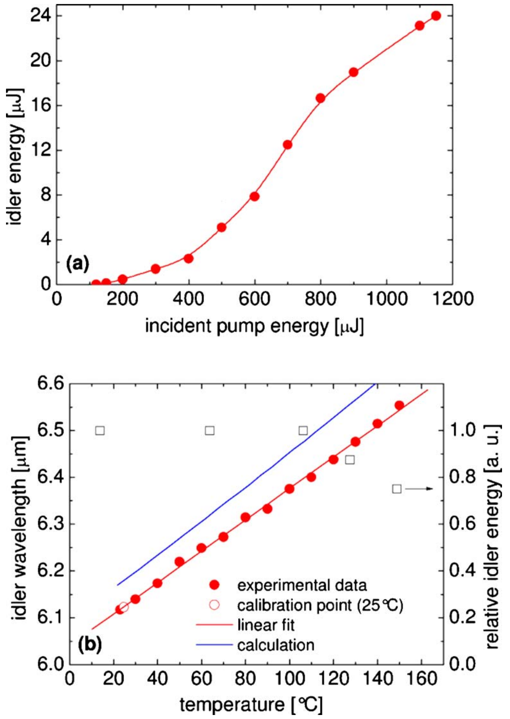

Fig. 2. (Color online) (a) Input–output characteristics of the CSP OPO at room temperature and (b) temperature tuning in non-critical configuration.

Temperature tuning was studied from room temperature to 150°C; see Fig. 2(b). To accommodate the oven, the cavity length had to be increased to 11 mm. All four lateral surfaces of the crystal were in good thermal contact with the metal holder, which was heated and where the temperature was measured by a calibrated sensor. The idler wavelengths were calculated from the signal wavelengths measured with a low resolution (~10 nm) InGaAs spectrometer (Avantes Model NIR256–1.7). Tuning from 6.117 to 6.554 μm was obtained for the idler wavelength. Thus CSP possesses nice tuning capability under noncritical conditions, which allows one to utilize its transparency up to the intrinsic limit set by multiphonon absorption. Note that in the well-known, but optically positive nonlinear crystal $ZnGeP_2$ that, as CSP, belongs to the II–IV–V_2 chalcopyrite class, the temperature tuning is very limited, e.g., 1 nm/deg for Type II noncritical configuration and 2.79 μm pump [8], which is obviously of no practical importance. From Fig. 2(b) it is clear that CSP is far more tunable than $ZnGeP_2$, and recent OPO experiments with critical phase-matching and 1.99 μm pump wavelength (Tm laser) show that the temperature tuning coefficient could be 1 order of magnitude (~10 nm/deg) higher than in $ZnGeP_2$ [9].

The calibration point at 25°C in Fig. 2(b) corresponds to measurement of the frequency-doubled (in a 5 mm Type I BiB_3O_8 crystal) signal by a high-resolution (0.25 nm) visible spectrometer. The calibration of this spectrometer (Acton Model SpectraPro 2150i) was confirmed by a He–Ne laser whose wavelength is very close to the second harmonic measured. The upper line in Fig. 2(b) shows a calculation based on extrapolation of recently obtained temperature-dependent Sellmeier equations, derived from refractive index measurements in the 95–295 K range [9]. The deviations in Fig. 2(b) amount to

60…100 nm for the idler wavelength, and in terms of temperature they are of the order of 20 K. However, at the signal wavelength, the discrepancy between experiment and calculation is only 3…3.5 nm.

The output idler energy is almost constant when changing the temperature, slightly decreasing above 6.4 μm, partially due to the idler absorption [Fig. 1(a)], to 75% of its maximum value, toward the longest idler wavelength achieved; see Fig. 2(b).

The OPO linewidth was measured at the signal wavelength (1.288 μm) using a 1-mm-thick Ag-coated CaF_2 Fabry–Perot etalon. We obtained $\sim$54 GHz or 1.8 cm^{-1} which is roughly 3.5 times smaller than the spectral acceptance calculated for the three-wave nonlinear process.

The signal-pulse duration, measured by a fast (70 ps response) InGaAs photodiode, amounted to 0.75 ns, shorter, as expected, than the pump-pulse duration of 1 ns (FWHM). Both pulse profiles are shown in Fig. 3. Unfortunately, the fastest HgCdTe detectors operating in the idler wavelength range have a response time $>$1 ns (Vigo System, Poland). On the other hand such pulse durations are too long for a practical autocorrelator setup. Nevertheless, there is no doubt that the idler pulse duration will be similar to the signal pulse duration because the pulse-shortening phenomenon in such an OPO, as a result of the gain-narrowing effect, is a very well-known fact confirmed in many experiments [10].

No crystal surface damage occurred in the present OPO which was pumped up to peak on-axis intensity of 32 MW/cm^2 at 1064 nm. This is higher than the damage level observed for the same kind of coating when using longer pulses (14 ns) [4]. The latter can be considered as an evidence for a further advantage of pumping with shorter pulses, all this possible, however, only owing to the high nonlinearity of CSP.

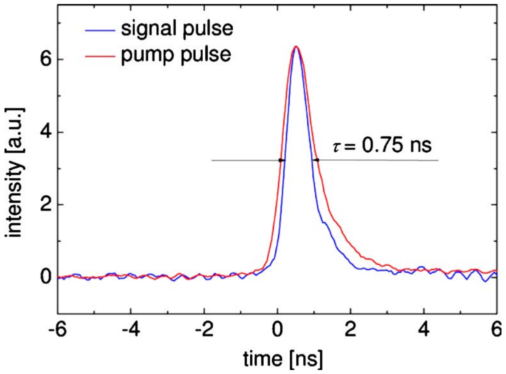

Fig. 3. (Color online) Signal pulse profile (blue curve) in comparison with the pump pulse (red curve) showing the pulse shortening effect and the subnanosecond pulse durations achieved.

In conclusion, we demonstrated for the first time to our knowledge OPO operation of 1064 nm pumped CSP at kilohertz repetition rate and subnanosecond signal and idler pulse durations achieving record average powers for a chalcopyrite material in the mid-IR. Temperature tuning is possible at elevated temperatures, and the idler covers the 6.117 to 6.554 μm range at 90° phase matching. Future work will be focused on reduction of the residual loss of CSP, while energy and power scaling will require the development of what we believe to be novel pump sources capable of generating $\sim$1 ns pulses at 1064 nm at higher power level. The present study showed that such special pump sources help avoid damage related problems.

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