Noncritical singly resonant optical parametric oscillator operation near 6.2 μm based on a $CdSiP_2$ crystal pumped at 1064 nm

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$CdSiP_2$ is employed in a nanosecond, 90°-phase-matched, singly resonant optical parametric oscillator pumped at 1064 nm to produce idler pulses near 6.2 μm with an energy as high as 470 μJ at 10 Hz.

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Owing to the lack of solid-state lasers, the spectral range above 3 μm in the mid-IR can be continuously covered only by nonlinear frequency downconversion. Oxide crystals can be pumped by widely used high-power diode-pumped laser systems, such as Nd:YAG, and perform well up to 4 μm, but their performance at longer wavelengths is dramatically affected by the onset of multiphonon mid-IR absorption. On the other hand, since efficient frequency conversion is only possible using pulsed laser sources (femtosecond to nanosecond) most of the chalcogenide mid-IR crystals will suffer two-photon absorption (TPA) at the pump wavelength of 1064 nm because of their low bandgap. Recently, we compared the properties of all potential candidates that can be pumped near 1064 nm, taking into account the TPA, residual absorption, birefringence, effective nonlinearity, thermal conductivity, and limitations related to the growth, availability, and some optomechanical properties [1]. Operation in the nanosecond regime is free of restrictions related to the spectral acceptance or higher-order nonlinear effects and has the best potential for achieving high average power and single-pulse energy. Nanosecond optical parametric oscillators (OPOs), pumped in the 1 μm range, have been demonstrated, however, with only 5 of the 14 compounds analyzed in [1]: Ag_3AsS_3 [2–4], $AgGaS_2$ [5–8], $HgGa_2S_4$ [9], $LiInSe_2$ [10], and the solid solution $Cd_xHg_{1-x}Ga_2S_4$ [11]. Apart from the archive Ag_3AsS_3 [4], oscillation at idler wavelengths exceeding 4.4 μm has been demonstrated only with $AgGaS_2$ [7,8], achieving impressive tunability from 3.9 to 11.3 μm [7].

The recently discovered cadmium silicon phosphide, $CdSiP_2$ (CSP) [12], is a negative uniaxial II–IV–V_2 chalcopyrite compound (space group 42 m) that allows 1064 nm pumping without TPA with a useful transparency up to 6.5 μm, limited by intrinsic multiphonon peaks. As shown in [1] it outperforms all other materials in almost every aspect relevant to high energy/average power generation (see above), 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 $d_{\mathrm{eff}}=d_{36}=84.5$ pm/V [13]. Here, we demonstrate what we believe to be the first OPO based on this material 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 8 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 0.198 cm^{-1} at 1064 nm, 0.114 cm^{-1} near 1.3 μm, and 0.014 cm^{-1} near 6.2 μm. Both faces were antireflection (AR) coated for the three wavelengths (pump, signal, and idler), and the eight-layer coating (Twin-Star) resulted in averaged reflectivities per surface of ~0.35% at 1.064 μm, ~0.4% at 1.285 μm, and ~0.5%

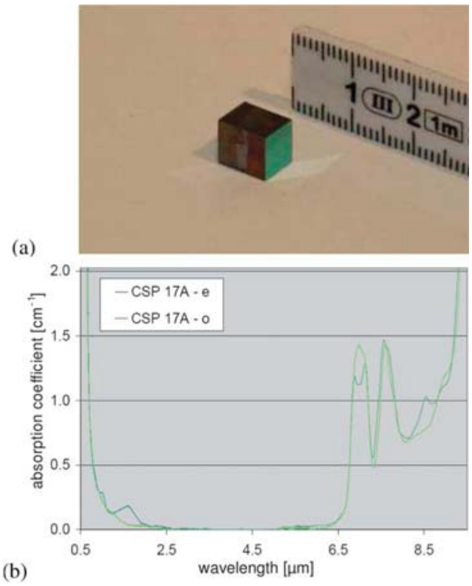

Fig. 1. (Color online) (a) AR-coated CSP 17A sample used and (b) its polarized transmission measured prior to coating.

at 6.2 μm.

The OPO cavity used is shown in Fig. 2. It consisted of two plane mirrors with a separation of 9.5 mm. The rear total reflector (TR) was an Ag-mirror (Balzers) with a reflection of $>98.5\%$ at the pump, signal, and idler wavelengths. 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. However, the signal was not totally reflected by the output coupler to avoid extreme intracavity fluence that could damage the crystal. The CSP crystal was pumped through the output mirror, which had a transmission of 82% at 1064 nm. The beams were separated by the pump bending mirror, (BM), which had high reflection for the pump ($R=98\%$ for p polarization) and transmitted 37% and 64% (s polarization) at the signal and idler wavelengths, respectively. Both the plane-parallel output coupler and the bending mirror were on ZnSe substrates with uncoated rear surfaces.

The pump source was a diode-pumped and electro-optically Q-switched Nd:YAG laser (Innolas) optimized for a repetition rate of 100 Hz. According to the specifications, its linewidth amounts to 1 cm^{-1}, M^2 is <1.5, and the divergence is <0.5 mrad. The laser generated 100 mJ, 14 ns (FWHM) pulses with an average power of 10 W. The measured energy stability was ±1%. A mechanical shutter (S) with an aperture of 8 mm, operating up to 50 Hz (nmLaser), was employed to reduce the repetition rate and thus the average pump power. A combination of a half-wave plate ($\lambda/2$) and a polarizer (P) served to adjust the pump energy. The pump laser was protected by a Faraday isolator, and the separation to the OPO was large enough to avoid feedback during the Q-switching process. The pump beam was not focused and had a Gaussian waist of $w\sim 1.9$ mm in the position of the OPO. The output of the OPO, behind the bending mirror, was detected by a calibrated pyroelectric energy meter positioned in front of the focus of a 10 cm MgF_2 lens (L). Only the idler energy was measured; the residual pump radiation and the signal were blocked by a 2.5 μm cut-on filter, F.

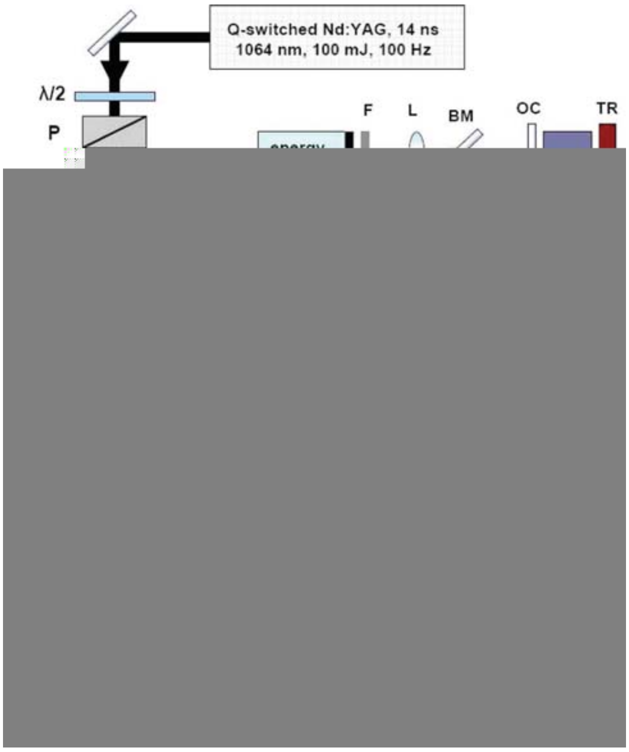

Fig. 2. (Color online) (a) Experimental setup and (b) photograph of the compact OPO. $\lambda/2$, half-wave plate; P, polarizer; S, mechanical shutter; F, 2.5 μm cut-on filter; L, 10 cm MgF_2 lens; D, diaphragm; BM, bending mirror; OC, output coupler; TR, total reflector.

Only normal incidence was studied in the present configuration, since the cavity was as short as possible in order to reduce the OPO threshold. In this noncritical scheme, the measured signal wavelength was 1.285 μm, corresponding to an idler wavelength of 6.193 μm. The calculation using the refined Sellmeier equations [13] gives an idler wavelength of 6.18 μm, but this slight deviation corresponds to only 0.6 nm at the signal wavelength, which is below the accuracy of the spectrometer. This is a good confirmation for the reliability of the present Sellmeier equations. The duration of the signal pulse, measured by a fast InGaAs photodiode, was 10 ns.

The pump threshold was about 1.8 mJ (~ 16 mJ/cm^2). The threshold can be calculated by using Brosnan and Byer's formula [14] for a singly resonant OPO with recycled pump. We used the exact experimental parameters, correcting for the pump beam absorption after the first pass and assuming equal (averaged for signal and idler) absorption of 0.064 cm^{-1} for the resonated wave. The nonlinear coefficient d_{36} of CSP was rescaled using Miller's rule, which gives an effective nonlinearity of $d_{\text{eff}}=92.3$ pm/V for this process [1]. The result for the threshold pump fluence was 29 mJ/cm^2. This value correlates better with the experimental peak (on-axial) fluence (~ 32 mJ/cm^2), which can be explained by the fact that oscillation starts in the central part of the pump beam. Then the estimated threshold in terms of energy would be 1.65 mJ. Note that the fact that the idler is also resonated to some extent (100% reflected by the rear mirror and 25% reflected by the output coupler) is not taken into account in that model.

The maximum idler energy measured at 10 Hz repetition rate was 0.47 mJ, at an incident pump energy of 21.4 mJ. This gives a conversion efficiency of 2.2% for the idler alone or a quantum conversion efficiency of $\sim 12.8\%$. Only slightly lower output powers were observed at 20 Hz, which can be attributed to the residual crystal absorption at the pump wavelength. The maximum average output power (idler only), reached in this case, was 9.1 mW. The measurements in Fig. 3 extend to an upper limit, where surface damage to the AR coating of the input face was observed. In terms of average fluence (~ 0.22 J/cm^2), the damage threshold is similar to that reported for $AgGaS_2$ [7]. Since the expectations for CSP are much higher [1], we believe the present damage is related

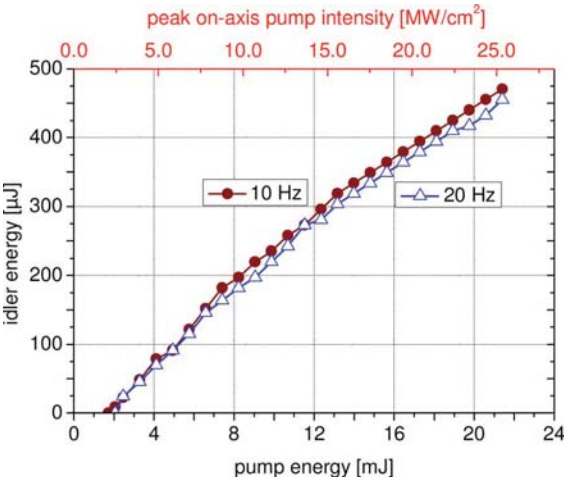

Fig. 3. (Color online) Idler output energy versus incident pump energy on the OPO crystal for two repetition rates.

to the AR coating. Single-layer coating is believed to solve this problem in the future. Nevertheless, the output energy level achieved with this very first sample of CSP already exceeds the best result previously reported at such long wavelengths with $\sim 1\ \mu m$ pumped OPOs, namely $372\ \mu J$ at $6\ \mu m$ using $AgGaS_2$ [7]. Moreover, the input/output characteristics in Fig. 3 show no saturation, in contrast to Fig. 4 in [7], which means that power scaling can be expected even without increasing the pump beam diameter.

The OPO linewidth, measured at the signal wavelength using a 1-mm-thick Ag-coated CaF_2 Fabry–Pérot etalon, was ~ 52 GHz ($\sim 1.7\ cm^{-1}$). This is roughly 3.5 times less than the spectral acceptance for the three-wave nonlinear process. The pulse-to-pulse stability for the idler pulses measured at an output level of $350\ \mu J$ was ±5%.

Damage tests were performed also extracavity, using 2 mm plates with the same AR coating only on the rear side. Damage to this surface developed (from dots to a crack) between 0.4 and $0.5\ J/cm^2$ (average fluence). Full damage of the front (uncoated) surface occurred at $\sim 10\%$ higher levels, but blackening started already from about $0.1\ J/cm^2$. It seems the AR coating helps to avoid the latter effect, but the question of whether the damage threshold is indeed lower in the presence of the signal wave (intracavity) still has no satisfactory answer.

In conclusion, we demonstrated for the first time to our knowledge OPO operation of CSP pumped at 1064 nm. Future work will be focused on reduction of the residual loss, improvement of the crystal surface damage resistivity, new cavity designs for maximum extraction of the idler energy, temperature tuning maintaining the noncritical configuration, and power scaling using crystals of larger aperture.

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