Non-critical singly resonant OPO operation near 6.2 µm based on a $CdSiP_2$ crystal pumped at 1064 nm

Peter G. Schunemann, Kevin T. Zawilski, Thomas M. Pollak
BAE Systems, Inc., MER15-1813, P.O. Box 868, Nashua, NH 03061-0868, USA, Tel. 603-885-5041, Fax. 603-885-0207,
peter.g.schunemann@baesystems.com

Valentin Petrov
Max-Born-Institute for Nonlinear Optics and Ultrafast Spectroscopy, 2A Max-Born-Str.,
D-12489 Berlin, Germany, Tel.: 49-30-63921272, Fax: 49-30-63921289, *petrov@mbi-berlin.de*

Abstract: $CdSiP_2$ 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.

OCIS Codes: (190.4400) Nonlinear optics, materials; (190.4970) Parametric oscillators and amplifiers

Due 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 down-conversion. 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 multi-phonon 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 band-gap. 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 opto-mechanical 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 OPOs, pumped in the 1 µm range, have been demonstrated, however, only with 5 of the 14 compounds analyzed in [1]: Ag_3AsS_3, $AgGaS_2$, $HgGa_2S_4$, $LiInSe_2$, and $Cd_xHg_{1-x}Ga_2S_4$. Apart from the archive Ag_3AsS_3 [2], oscillation at idler wavelengths exceeding 4.4 µm has been demonstrated only with $AgGaS_2$ [3], achieving impressive tunability from 3.9 to 11.3 µm.

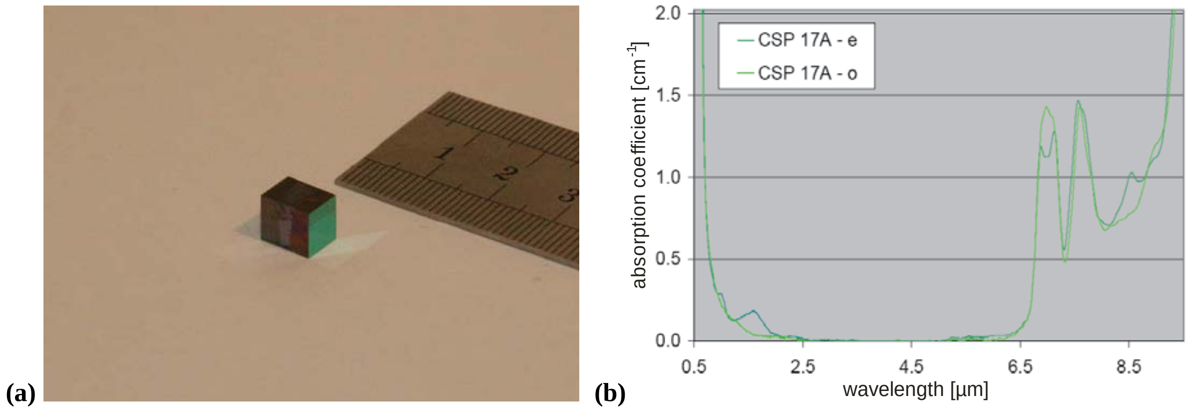

Fig. 1. The AR-coated CSP 17A sample used (a) and its polarized transmission measured prior to coating (b).

The recently discovered cadmium silicon phosphide, $CdSiP_2$ (CSP) [4], is a negative uniaxial II-IV-V_2 chalcopyrite compound (space group 42m) that allows 1064 nm pumping without TPA with a useful transparency up to 6.5 µm, limited by intrinsic multi-phonon 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 band-gap which is not intrinsic. In addition, it is the only material which, without being a solid solution, still allows non-critical phase-matching with a maximum effective nonlinearity of $d_{eff}=d_{36}$=84.5 pm/V [5]. Here, we demonstrate 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 (HGF) technique. It was cut at θ=90°, φ=45° and had a length of 8 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 0.198 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 AR-coated for the three wavelengths (pump, signal, and idler) and the 8-layer coating (TwinStar) resulted in averaged reflectivities per surface of ~0.35% at 1.064 µm, ~0.4% at 1.285 µm and ~0.5% 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, OC, and the bending mirror, BM, were on ZnSe substrates with uncoated rear surfaces.

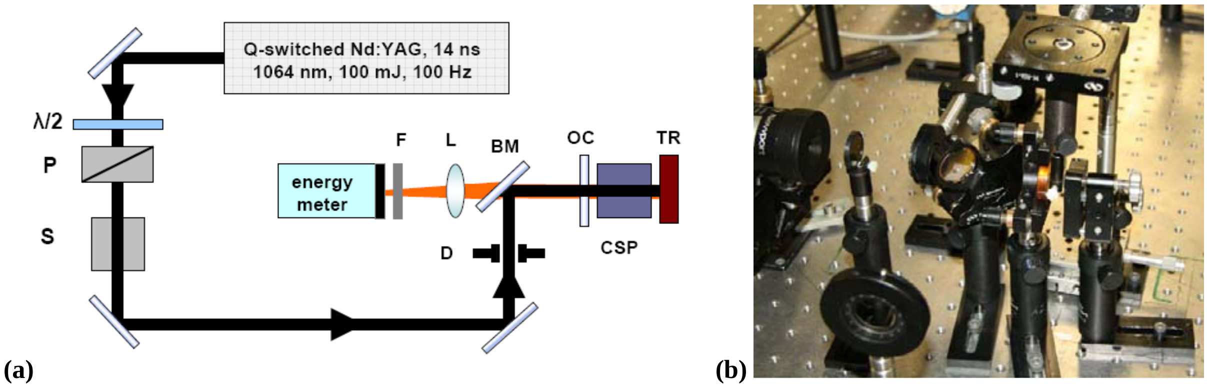

Fig. 2. Experimental set-up (a) and photograph (b) of the compact OPO. $\lambda/2$: half-wave plate, P: polarizer, S: mechanical shutter, F: 2.5 µm cut-on filter, L: 10 cm MgF_2 lens, D: diaphragm, BM: bending mirror, OC: output coupler, TR: total reflector.

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~1.9 mm in the position of the OPO. The output of the OPO, behind the bending mirror, BM, 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.

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 non-critical scheme, the measured signal wavelength was 1.285 µm, corresponding to an idler wavelength of 6.187 µm. The calculation using the refined Sellmeier equations [5] gives an idler wavelength of 6.18 µm but this slight deviation corresponds to only 0.5 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 & Byer's formula [6] 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_{eff}=92.3 pm/V for this process [1]. The result for the threshold pump energy was 2.4 mJ. The somewhat lower threshold measured in the experiment can be explained by the fact that the idler is also resonated to some extent (100% reflected by the rear mirror and 25% reflected by the output coupler).

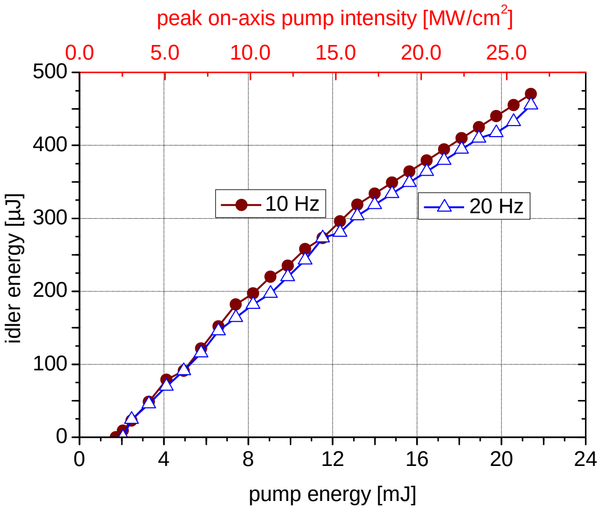

Fig. 3. Idler output energy versus incident pump energy on the OPO crystal for two repetition rates.

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 ~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 fluence (~0.19 J/cm^2), the damage threshold is similar to that reported for $AgGaS_2$ [3]. Since the expectations for CSP are much higher [1], we believe the present damage is related 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 ~1 µm pumped OPOs, namely 372 µJ at 6 µm using $AgGaS_2$ [3]. Moreover, the input/output characteristics in Fig. 3 show no saturation, in contrast to Fig. 4 in Ref. [3], 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-Perot etalon, was ~45 GHz (~1.5 cm^{-1}). The pulse-to-pulse stability for the idler pulses measured at an output level of 350 µJ was +-5%.

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 non-critical configuration, and power scaling using crystals of larger aperture.

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