Sub-nanosecond, 1-kHz, temperature-tuned, non-critical mid-IR OPO based on $CdSiP_2$ crystal pumped at 1064 nm

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Abstract: Temperature tuning (6.117-6.554 µm for the idler) and sub-nanosecond durations are demonstrated with a non-critical, 1064-nm pumped $CdSiP_2$-OPO. At 1 kHz, the output idler energy of 24 µJ corresponds to an average power of 24 mW.

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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 multi-phonon 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 acceptance or higher order nonlinear effects, typical for ultrashort laser pulses, but even in that case the nonlinear crystals should possess a sufficiently large band-gap 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 multi-phonon 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 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 [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 near 6.2 µm with an energy as high as 470 µJ, at a repetition rate of 10-20 Hz.

In this work 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, sub-nanosecond signal and idler pulse durations, (ii) the good thermo-mechanical properties enabled for the first time to our knowledge 1-kHz repetition rate operation with a non-oxide nonlinear material (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 pumped at 1064 nm 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 non-critical tuning range to the long wavelength transmission limit, which covers the essential for medical applications spectral region near 6.45 µm.

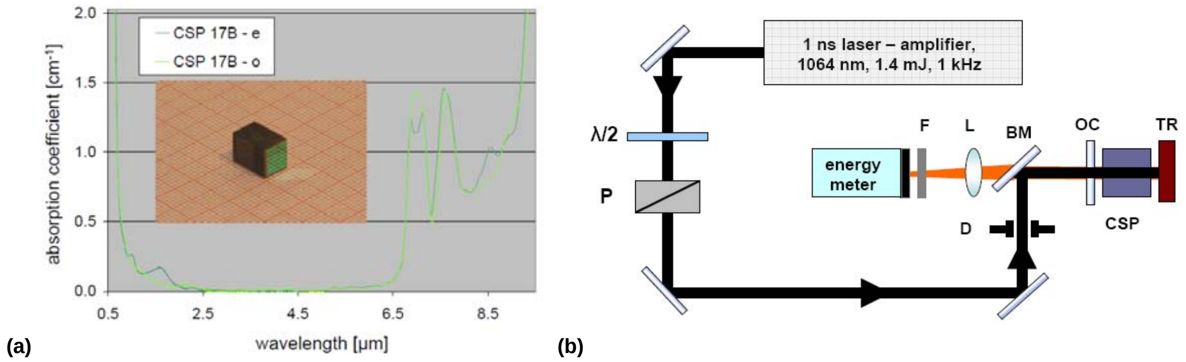

Fig. 1. The polarized transmission of the CSP 17B sample used, measured prior to coating, and a photograph of the AR-coated sample (inset), (a). Experimental set-up of the CSP OPO (b): $\lambda/2$: half-wave plate, P: polarizer, F: 5 µm cut-on filter, L: 10 cm MgF_2 lens, D: diaphragm, BM: bending mirror,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

The sample used in the present study (Fig. 1a, inset) was cut at θ=90°, φ=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 6.2-6.4 µm. Both faces were AR-coated for the three wavelengths (pump, signal, and idler) and the 8-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, however, slightly lower transmission of 77% at 1064 nm.

The OPO cavity was the same as the one used in [4], Fig. 1b. It consisted of two plane mirrors with a separation of 10 mm. The rear total reflector, TR, was an Ag-mirror with a reflection 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 mirror, BM, which had high reflection 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, a cw pumped Nd:YVO$_4$ regenerative amplifier, and a double pass Nd:YAG post amplifier with pulsed pumping, 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~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.

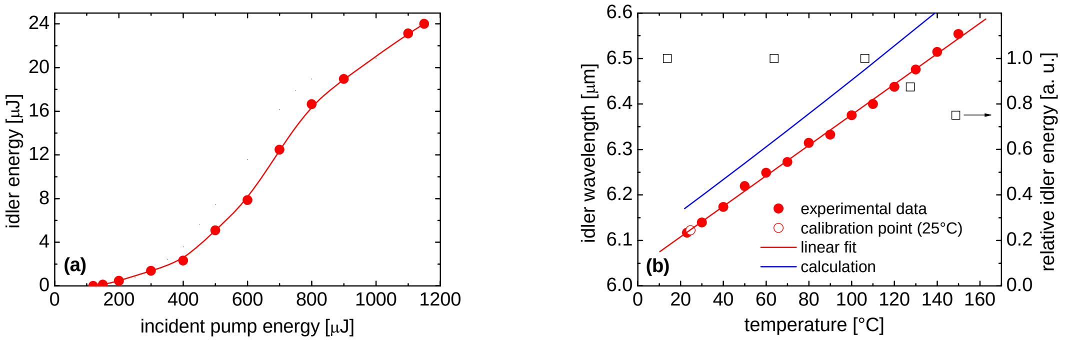

Fig. 2. Input output characteristics of the CSP OPO at room temperature (a) and temperature tuning in non-critical configuration (b).

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. 2a. At 25°C, maximum idler energy of 24 µJ was achieved at 6.125 µm. The OPO linewidth, measured at the signal wavelength (1288 nm) using a 1-mm-thick Ag-coated CaF$_2$ Fabry-Perot etalon, was ~54 GHz (1.8 cm^{-1}). The signal pulse duration, measured by a fast InGaAs photodiode amounted to 0.75 ns, shorter, as expected, than the pump pulse duration. No such fast MCT photodetectors exist for the mid-IR but the idler pulse duration should be very similar. The temperature tuning curve, obtained from the measured signal wavelength, is shown in Fig. 2b. To accommodate the oven, the cavity length had to be increased to 11 mm. The output idler energy is almost constant, slightly decreasing above 6.4 µm, partially due to the idler absorption (Fig. 1a), to 75% of its maximum value at the longest idler wavelength (6.554 µm) achieved. The calibration point at 25°C corresponds to measurement of the frequency doubled signal by a high resolution visible spectrometer. The blue line in Fig. 2b shows a calculation based on extrapolation of recently derived temperature-dependent Sellmeier equations fitted in the 95-295 K range.

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