Optical parametric generation in $CdSiP_2$ at 6.125 μm pumped by 8 ns long pulses at 1064 nm

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A 21.4 mm long noncritically cut $CdSiP_2$ crystal, pumped by 8 ns pulses at 1064 nm in a double-pass configuration for pump, signal, and idler, generated 523 µJ, 5.8 ns idler pulses at 6.125 µm. The average power of 52.3 mW at the repetition rate of 100 Hz is the highest ever achieved at such wavelengths with direct down conversion from the 1 µm spectral range.

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Efficient parametric frequency down-conversion relies either on optical parametric oscillators (OPOs) employing a resonant cavity or on optical parametric generators (OPGs) employing a single-pass traveling-wave scheme. A cavity is in general necessary for low peak pump powers, normally associated with pulses of duration from a few nanoseconds to cw, in which case many round-trips of the resonated wave (signal, idler, or both) ensure a sufficiently high parametric gain to reach threshold [1]. Synchronously pumped OPOs (SPOPOs) also rely on a cavity with a length matched to the cavity length of the ultrafast pump laser to reduce the threshold when using a high repetition rate (~100 MHz) but low peak power femtosecond or picosecond pump pulses [1]. Single-pass traveling-wave type OPGs, on the other hand, could only reach threshold with the high peak power available from ultrafast (femtosecond or picosecond, up to ~1 ns) amplified pump sources operating at lower repetition rates for which an OPO or SPOPO cavity would be impractical (1 ns corresponds to 30 cm in free space) [2].

The differentiation described above became less clear, however, with the development of periodically poled materials that provide substantially higher effective nonlinearities and thus require lower pump powers. Thus, e.g., an OPG based on 55 mm long, periodically poled $LiNbO_3$ (PPLN) could be pumped by 8.5 ns long pulses at 1064 nm [3]. Using periodically poled $KTiOPO_4$ (PPKTP) with 2.3 ns pump pulses at 1064 nm resulted in ~1 ns long signal pulses, both with a 10 mm long crystal in an OPO cavity (with only very few round-trips) and with a 20 mm long crystal in quasi-OPG (low-finesse doubly resonant OPO cavity parasitically formed by residual
1 reflections of the crystal AR coatings) [4].

Therefore, it can be expected that, in general, for pulse durations between 1 and 10 ns, high-gain nonlinear materials can be employed both in OPO and OPG configurations. The advantage of OPGs is that seeding is much easier to apply for narrow-band single-frequency operation [3] and high spectral resolution spectroscopy.

Here we demonstrate that such regimes are possible with the new nonlinear crystal $CdSiP_2$ (CSP) that possesses extremely high (84.5 pm/V) second-order nonlinear coefficient [5] for birefringent phase-matching, resulting in a figure of merit ($\mathrm{FOM} = d^2/n^3$ where d is the effective nonlinearity and n is an average refractive index), that is ~7 times higher than that of $HgGa_2S_4$ [6], the best previously known material that could be pumped at 1064 nm without two-photon absorption losses. The transparency of CSP permits idler tuning up to ~6.5 μm in the mid-IR. We have already achieved sub-ns OPO operation (both at the signal and idler wavelengths) using 1 ns long 1064 nm pulses to pump a CSP-based OPO [7], which represents a unique OPO regime in terms of pulse durations. In that case, a small but sufficient number of round-trips was possible due to the very short crystal (9.5 mm) and cavity (10 mm) lengths. In the present work we employ for the first time a long noncritically cut CSP crystal in a double-pass OPG configuration that showed extremely low threshold even for pump pulse duration as long as 8 ns at 1064 nm. This in turn represents a unique OPG regime that is of practical importance because commercial *Q*-switched pump systems normally deliver pulses with duration of the order of 10 ns or longer.

The CSP crystal available for this experiment was grown at BAE Systems, Inc. It was 21.4 mm long with an aperture of 4.1 (along *c*-axis) × 6.1 mm² and cut for noncritical (90°) type-I (oo-e) interaction (Fig. 1). Both faces were AR coated with a single layer of Al_2O_3 (TwinStar), which in fact was optimized only for the pump and signal wavelengths (minimum reflection of ~1% at 1150 nm), hoping in this way to achieve higher surface damage threshold. The actual reflectivity measured at the three wavelengths was 1.3% (1064 nm), 2% (1288 nm), and 20% (6.125 μm).

The pump beam from a diode-pumped *Q*-switched Nd:YAG laser/amplifier system operating at 100 Hz (Innolas), after spatial filtering and attenuation by a system of wave plate and polarizer, was expanded to a slightly elliptical shape with approximately Gaussian spatial distribution

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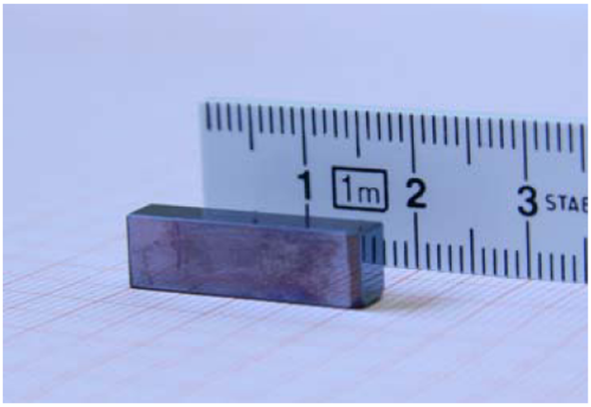

Fig. 1. (Color online) Photograph of the AR-coated CSP sample.

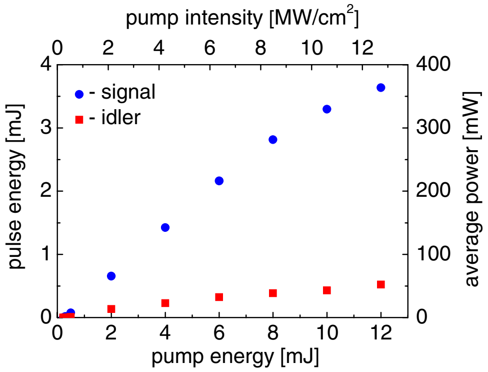

Fig. 3. (Color online) Signal (1288 nm) and idler (6.125 μm) output energy, and average power at 100 Hz versus incident pump energy at 1064 nm.

and diameter of ~10.4 and ~12.1 mm in the horizontal and vertical (along the crystal ***c***-axis) directions, respectively. A nearly flat-top spatial profile was then obtained by a circular aperture, which reduced the beam diameter to ~3.8 mm, matching the limited crystal aperture. The pump pulse duration was 8 ns.

The CSP crystal was pumped in double-pass using a 45° ZnSe bending mirror for the pump radiation, which was highly transmitting at both signal and idler wavelengths, and a metal (Ag, $R > 98\%$) mirror to retroreflect all the three pulses for a second pass (see Fig. 2). All separations were kept as short as possible to avoid air absorption of the idler, which is typically of the order of 0.5%/cm.

The OPG threshold was found at 213 μJ of incident pump energy (~0.23 MW/cm^2 peak on-axis intensity), as shown in Fig. 3. At the maximum applied pump energy of 12 mJ (12.7 MW/cm^2), the total output energy exceeded 4 mJ, from which ~3.64 mJ were at 1288 nm (signal) and ~0.52 mJ at 6.125 μm (idler). The fluctuations were ±5%, measured for the idler. There was some trend of saturation of the idler energy—the ratio of the signal to idler energy increases with the pump level reaching ~7 at maximum level, while the theoretical value (without taking into account the different reflection of the crystal AR coatings) should be around 4.8. There is still no explanation for this power dependent loss of idler energy.

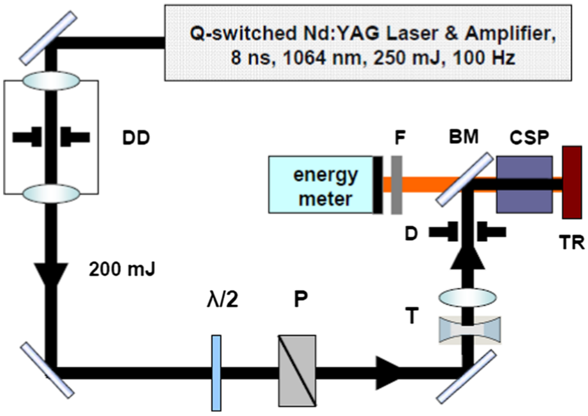

Fig. 2. (Color online) OPG setup. *T*: telescope, *D*: diaphragm, BM: bending mirror, TR: total reflector, *F*: exchangeable filters, *P*: polarizer, $\lambda/2$: half-wave plate, DD: diamond diaphragm for spatial profile cleaning.

The threshold obtained in terms of pump intensity is extremely low even keeping in mind the long pump pulse duration. The lowest OPO pump threshold in terms of pump energy we are aware of is 2 μJ at 3.1 μm for pumping a noncritically cut 24 mm long type-II ZnGeP$_2$ crystal, which has similarly high nonlinearity [8]. This OPO utilized double pump pass and only the signal was resonated. In terms of pump fluence and intensity for the 10 ns long pulses at 3.1 μm the 2 μJ threshold energy is equivalent to 8.2 mJ/cm^2 and 0.82 MW/cm^2, respectively. Thus, in fact, the threshold measured in the present work with CSP is roughly four times lower, 1.84 mJ/cm^2. Note that an OPG should in general exhibit a higher threshold than an OPO.

Obviously, residual reflections may contribute in our case to an OPO feedback effect, similar to what has been observed in [4] with PPKTP. We checked this by tilting the crystal in order to facilitate noncollinear interaction and the conclusion was that the surface reflections formed a low-finesse cavity for the idler. This could be expected since the AR coating was not optimized for the idler wavelength and the residual reflection was substantially higher than for the signal. Thus, the present experiment corresponds more or less to quasi-OPG or weakly resonant OPO operation. Since optimization of AR coatings for extremely low reflectivity at both signal and idler wavelengths requires multilayers, which are related to decreasing damage resistivity, better AR coatings are not expected to suppress totally the OPO effect. On the other hand, wedged sample, as in the case of the thin PPLN used in [3], is not expected to be effective either because the aperture of the CSP crystal is relatively large. Instead, we plan to realize a pure OPG experiment with this long CSP sample using shorter pump pulses of less than 600 ps duration, which nowadays are available from some specially designed diode-pumped Nd-laser systems [9]. Having in mind the large refractive index of CSP (~3.2) OPO feedback should not be possible for a crystal length exceeding 20 mm.

The temporal pulse profiles for the present setup were measured using fast photo-detectors and are shown in Fig. 4. As expected the signal and idler have shorter pulse durations of 4.4 and 5.8 ns, respectively, than the pump.

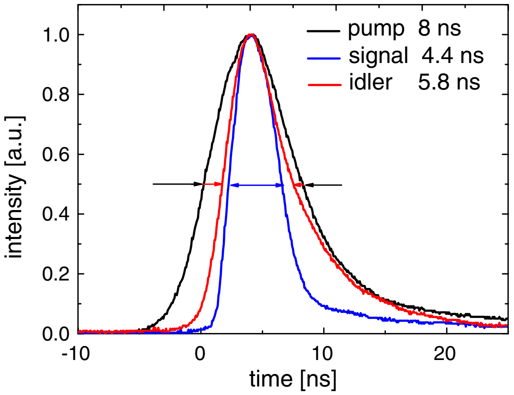

Fig. 4. (Color online) Pump, signal, and idler temporal shapes at maximum output energy. The recorded idler profile is possibly affected by the 2.6 ns response time of the HgCdTe photoconductive detector. The numbers indicate the FWHM.

The spatial properties of the OPO output beams were measured by a 10 cm MgF_2 focusing lens. Estimating the beam diameter with the knife-edge method gave M^2 values of 7.1 and 7.8 for the idler in the horizontal and vertical directions, respectively.

Taking into account the maximum signal pulse energy obtained with the present double-pass CSP-based OPG, we arrive at a quantum conversion efficiency of 34.7% (or ~25% if the idler is considered that experiences some losses). Before the onset of saturation, e.g., at a pump power of 6 mJ, these efficiencies amount to 41.4% and ~31%, respectively. All these values are much higher than the OPO quantum conversion efficiency achieved with a shorter crystal length of ~1 cm (this sample did not operate as OPG up to the highest pump level applied) that amounted to 12.8%, using for the calculation the idler output because the signal was resonated [10]. Moreover, maximum pump pulse energy and peak intensity were
2 about two times higher in [10] and so the risk of damage. We repeated the experiment from [10] at 100 Hz, with a pump beam profile shaped to a quasi-flat top in the same manner as in the present work, but the quantum conversion efficiency remained at ~13%, and the M^2 factor (~11) was even worse than in the present, to a greater extent OPG, configuration.

Therefore, we conclude that the OPG concept is feasible in the temporal regime between 1 and 10 ns for achieving higher output energies and average powers. This is especially true for highly nonlinear crystals that still exhibit residual absorption losses (at the pump and signal wavelengths) such as the CSP crystal that is still in the development stage. Nevertheless, it should be outlined that in comparison with all other nonoxide materials that have sufficiently large bandgap to be pumped near 1 μm, such as $AgGaS_2$, $HgGa_2S_4$, $LiInSe_2$, and isomorphs, or some mixed (solid solution) crystals [6], apart from its upper transmission limit, CSP is by far the best candidate for high power/high energy operation in the mid-IR based on direct (single stage) downconversion from the ~1 μm spectral range.

While, as already mentioned, realization of pure OPG operation would require special pump sources that are not commercially available, the presence of residual crystal surface reflections is not something that should be necessarily avoided as far as power/energy scaling is concerned. On the contrary, the issues with the resistivity of the AR-coatings for CSP are still not resolved and present data on their effect on the surface damage threshold of CSP are not conclusive. This means that uncoated crystals of CSP with plane-parallel faces could be used for resonant enhancement of the OPG regime, simultaneously avoiding the necessity of AR coatings, or one of the surfaces can be Au-coated, as previously demonstrated for $ZnGeP_2$ [8], for retro-reflection of the three waves. In any case, such designs are especially suited for CSP because it is also the only nonoxide material that enables noncritical phase-matching and temperature tuning [6] for pump wavelengths in the 1 μm spectral range.

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Queries

1. Does “AR” stand for “antireflection”?

2. could we add the word “was” after “so” for grammar?

3. Is Laser Science and Technology, An International Handbook the name of the book for Ref. 1 and 2?