

# Mid-Infrared Optical Parametric Generation in CdSiP<sub>2</sub> Crystal Pumped by 8-ns Long Pulses at 1064 nm

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**Abstract:** A 21.4-mm-long non-critically cut CdSiP<sub>2</sub> 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 and 100 Hz.

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## 1. Introduction

For decades there has been a clear differentiation between optical parametric oscillators (OPOs) and optical parametric generators (OPGs), not only because the former consist of a resonant cavity and the latter are of traveling-wave type but also with respect to the pulse durations. Cavity is in general necessary for low peak pump powers, normally associated with pulses of duration from few nanoseconds to continuous-wave in which case many round trips of the resonated wave (signal or idler, or both) ensure sufficiently high parametric gain to reach threshold. Traveling-wave type OPGs on the other hand could only reach threshold with the high peak power available from ultrashort (femtosecond or picosecond, up to ~1 ns) pump sources for which anyway a cavity would be impractical (1 ns corresponds to 30 cm in free space). This situation changed, however, with the development of periodically-poled materials which provide substantially higher effective nonlinearities and thus require lower pump powers. Thus, e.g. an OPG based on 55-mm long, periodically-poled LiNbO<sub>3</sub> (PPLN) could be pumped by 8.5-ns long pulses at 1064 nm [1]. Using periodically poled KTiOPO<sub>4</sub> (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 reflections of the crystal AR-coatings) [2]. Thus, it is clear that high-gain nonlinear materials can be applied for pulse durations of the order of 1 to 10 ns both in OPO and OPG configurations. The advantage of OPG is that seeding is much easier to apply for narrow-band single-frequency operation [1]. Here we demonstrate that such regimes are possible also with the new nonlinear crystal CdSiP<sub>2</sub> (CSP) which possesses extremely high (84.5 pm/V) second-order nonlinear coefficient [3] for birefringent phase-matching. This crystal, which can be pumped at 1064 nm without two-photon absorption, extends the idler wavelength range into the mid-IR up to about 6.5 μm. 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 [4]. 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 non-critically cut CSP crystal in a double-pass OPG configuration which showed extremely low threshold even for pump pulse duration as long as 8 ns at 1064 nm.

## 2. Experimental set-up

The CSP crystal available for this experiment was 21.4 mm long with an aperture of 4.1 (along *c*) × 6.1 mm<sup>2</sup>. It was cut for non-critical (90°) type-I (oo-e) interaction, Fig. 1a. Both faces were AR-coated with a single layer of Al<sub>2</sub>O<sub>3</sub> (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 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  $\sim 3.8$  mm, fitting the limited crystal aperture. The pump pulse duration was 8 ns.

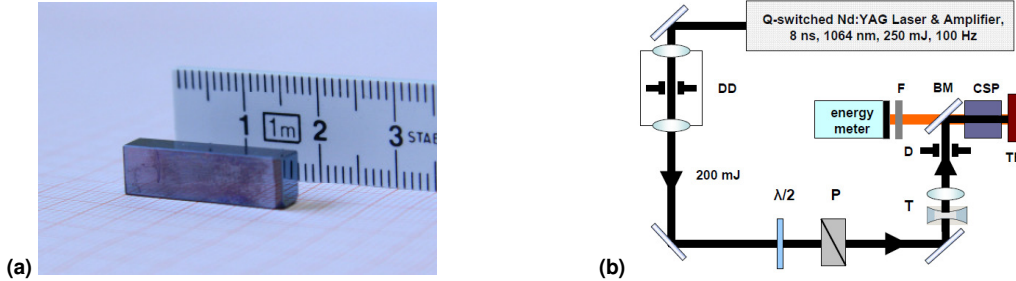


Fig. 1. Photograph of the AR-coated CSP sample (a) and OPG set-up (b). T: telescope, D: diaphragm, BM: bending mirror, TR: total reflector, F: exchangeable filters, P: polarizer,  $\lambda/2$ : half-wave plate, DD: diamond diaphragm for spatial profile cleaning.

The CSP crystal was pumped in double-pass using a  $45^\circ$  ZnSe bending mirror for the pump radiation which was highly transmitting at both the signal and idler wavelengths and a metal (Ag,  $R > 98\%$ ) mirror to retro-reflect all the three pulses for a second pass, see Fig. 1b. All separations were kept as short as possible to avoid air absorption of the idler which is typically of the order of  $0.5\%/cm$ .

### 3. Results and discussion

The OPG threshold was found at  $213 \mu J$  of incident pump energy ( $\sim 0.23 \text{ MW/cm}^2$  peak on-axis intensity), see Fig. 2. At the maximum applied pump energy of  $12 \text{ mJ}$  ( $12.7 \text{ MW/cm}^2$ ), the total output energy exceeded  $4 \text{ mJ}$ , from which  $3.635 \text{ mJ}$  were at  $1288 \text{ nm}$  (signal) and  $0.523 \text{ mJ}$  at  $6.125 \mu m$  (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  $\sim 7$  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.

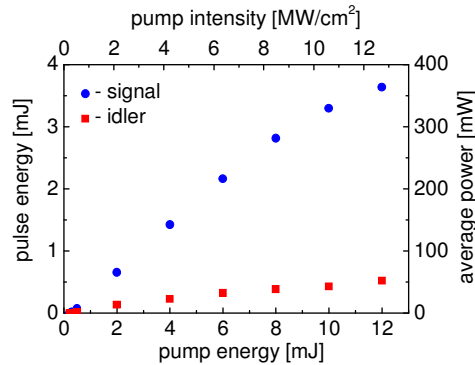


Fig. 2. Signal ( $1288 \text{ nm}$ ) and idler ( $6.125 \mu m$ ) output energy, and average power at  $100 \text{ Hz}$  versus incident pump energy at  $1064 \text{ nm}$ .

The threshold obtained in terms of pump intensity is extremely low even having in mind the long pump pulse duration. The lowest OPO pump threshold in terms of pump energy we are aware of, is  $2 \mu J$  at  $3.1 \mu m$  for pumping a non-critically cut  $24\text{-mm}$  long type-II  $\text{ZnGeP}_2$  crystal which has similarly high nonlinearity [5]. This OPO utilized double pump pass and only the signal was resonated. In terms of pump fluence and intensity for the  $10\text{-ns}$  long pulses at  $3.1 \mu m$  this gives, however,  $8.2 \text{ mJ/cm}^2$  and  $0.82 \text{ MW/cm}^2$ , respectively. Thus, in fact, the threshold measured in the present work with CSP is roughly four times lower,  $1.84 \text{ mJ/cm}^2$ . Obviously, residual reflections may contribute in our case to an OPO effect, similarly to what has been observed in [2] with PPKTP. We checked this by tilting the crystal in order to facilitate non-collinear 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-resonated OPO operation. Since optimization of AR-

coatings for extremely low reflectivity at both signal and idler wavelengths requires multi-layers which is 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 [1], 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 [6]. Having in mind the large refractive index of CSP ( $\sim 3.2$ ) OPO feedback should not be possible for a crystal length exceeding 20 mm.

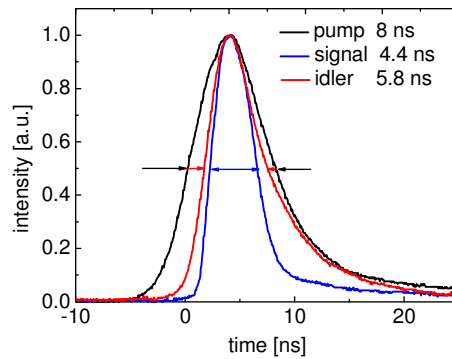


Fig. 3. Pump, signal and idler temporal shapes at maximum output energy. The recorded idler profile is possibly affected by the 2.6 ns response time of the HgCdTe photoconductive detector. The numbers indicate the FWHM.

The temporal pulse profiles for the present set-up were measured using fast photo-detectors and are shown in Fig. 3. As expected the signal and idler have shorter pulse durations of 4.4 and 5.8 ns, respectively, than the pump.

The spatial properties of the OPO output beams were measured by a 10-cm MgF<sub>2</sub> 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.

#### 4. Conclusion

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  $\sim 25\%$  if the idler is considered which experiences some losses). Before the onset of saturation, e.g. at a pump power of 6 mJ, these efficiencies amount to 41.4% and  $\sim 31\%$ , respectively. All these values are much higher than the OPO quantum efficiency achieved with a shorter crystal length of  $\sim 1$  cm (to avoid OPG effects) with which we obtained an efficiency of 12.8% using for the calculation the idler output because the signal was resonated [7]. Moreover, maximum pump pulse energy and peak intensity were about two times higher in [7] and so the risk of damage. We repeated the experiment from [7] 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  $\sim 13\%$  and the  $M^2$  factor ( $\sim 11$ ) was even worse. Thus, we conclude that the OPG concept is feasible in this temporal regime for achieving higher output energies and average powers with the highly nonlinear but still exhibiting residual absorption losses (at the pump and signal wavelengths) CSP crystal in the mid-IR starting from  $\sim 1$   $\mu$ m pump laser systems.

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