High-power, mid-IR HgGa2S4 OPOs pumped at 1064 nm

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High-power, mid-IR $HgGa_2S_4$ OPOs pumped at 1064 nm

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Abstract: $HgGa_2S_4$ is employed in 1064-nm pumped OPOs, to generate 5 to 7-ns long idler pulses with energies as high as 6.1 mJ near 4 µm and 3 mJ near 6.3 µm at 10-100 Hz.

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

Defect chalcopyrite nonlinear optical crystals with chemical formula $A^{II}B^{III}{}_2C^{VI}{}_4$ and point group $\bar{4}$ symmetry exhibit substantially higher second order nonlinear susceptibility compared to their chalcopyrite analogues but in fact only $HgGa_2S_4$ (HGS) has been used so far for phase-matched nonlinear frequency conversion [1]. Compared to the commercially available chalcopyrite $AgGaS_2$ (AGS, the standard non-oxide mid-IR material when it comes to pumping down-conversion schemes near 1 µm) HGS exhibits ~1.8 times higher nonlinear coefficient d_{36} and this at slightly increased band-gap value (2.79 eV for HGS against 2.7 eV for AGS), i.e. at somewhat improved damage resistivity [1]. Due to its wide band-gap, HGS, similar to AGS, can be pumped near 1 µm by short or ultrashort pulse laser sources (e.g. Nd:YAG at 1064 nm) without two-photon absorption. Only few other non-oxide crystals possess this property but most of them (the biaxial $LiGaS_2$, $LiInS_2$, $LiGaSe_2$, $LiInSe_2$, $BaGa_4S_7$) exhibit nonlinear susceptibility lower than AGS. The recently developed chalcopyrite $CdSiP_2$ (CSP) is highly nonlinear and non-critically phase-matchable crystal but transparent only up to ~6.5 µm [2]. Here we compare the properties of such nonlinear crystals and examine the potential of HGS in high-power OPOs with 1064 nm pumping achieving almost 10-fold increase in the idler average power with 610 mW (6.1 mJ at 100 Hz) near 4 µm and almost 6-fold increase in terms of energy at 6.3 µm (3 mJ) in damage free operation, with respect to the state of the art of such OPOs.

The HGS samples used were a 13.4 mm-long one with an aperture of ~10×13.6 mm^2 cut at θ=52.7° and φ=45° for type-I (oo-e) interaction for the 4-µm range (HGS-1), and a 10.76 mm-long one with an aperture of ~9.5×9.5 mm^2, cut at θ=50.2° and φ=0° for type-II (eo-e) interaction for the 6.3 µm range (HGS-2), in both cases utilizing only the d_{36} component of the nonlinear tensor. Effective nonlinearity is ~36% higher for type-II interaction. Both crystals were AR-coated for the resonated signal wavelength. A diode-pumped Q-switched Nd:YAG laser/amplifier system served as a pump, see Fig. 1a, with a bandwidth of 30 GHz (1 cm^{-1}) and M^2 factor of ~1.4. The pump beam reached the HGS crystals after reflection at the ZnSe bending mirror (M2) and passing through the output coupler (OC) which is HT for the idler and had a reflectivity ~70% at the signal wavelength.

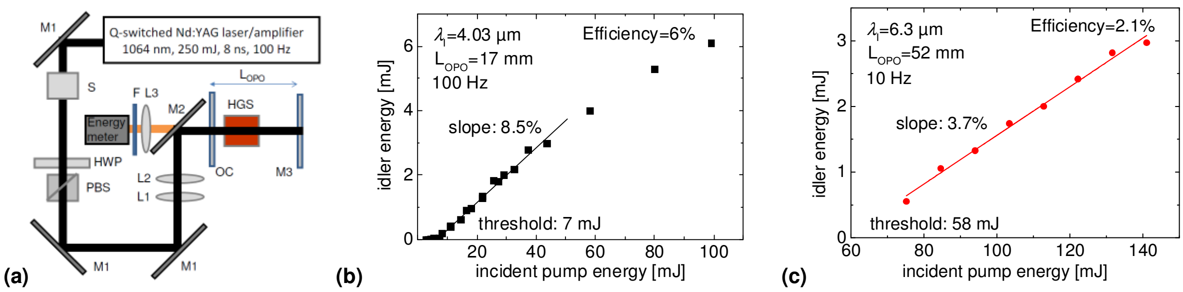

Fig. 1. HGS OPO experimental set-up (a). A mechanical shutter (S) can reduce the repetition rate and the average pump power, a half-wave plate (HWP) and a polarizing beam splitter (PBS) are used as attenuator and a telescope consisting of two lenses L1 and L2 expands the pump beam to a diameter of 9.6 and 8.45 mm in the horizontal and vertical directions, respectively. A plane Ag-mirror acts as a total reflector (M3) for all three waves in a double pump pass singly resonant OPO configuration. The filters (F) suppress the residual pump and signal pulses. Input-output characteristics with HGS-1 (b) and HGS-2 (c) with thresholds extrapolated from the idler slope efficiency.

Figures 1b,c show the input-output characteristics of the two crystals at normal incidence. The maximum pump level was kept <40 MW/cm^2 for HGS-1 and <55 MW/cm^2 for HGS-2 for which in addition the repetition rate was reduced to 10 Hz. The extrapolated threshold with HGS-1 corresponds to an axial pump fluence of 22 mJ/cm^2 or a peak intensity of ~2.7 MW/cm^2. The highest idler energy of 6.1 mJ obtained at 4.03 µm translates into an average power of 610 mW, the highest ever achieved with a 1-µm pumped OPO based on a non-oxide material [3]. The threshold with HGS-2 corresponds to an axial pump fluence of 180 mJ/cm^2 or a peak intensity of ~23 MW/cm^2, i.e. it is ~8-times higher although type-II interaction compensates for the reduced parametric gain. The increased threshold is partly due to the longer cavity but with L_{OPO}=20 mm the HGS-2 OPO did not operate better in terms of maximum idler energy because the slope efficiency was also lower. The highest idler energy of 3 mJ obtained at 6.3 µm with HGS-2 represents an improvement of >6 times compared to previous best results with CSP and $BaGa_4S_7$ based OPOs [2,4] and the highest idler energy ever demonstrated above 6 µm with a 1-µm pumped OPO based on a non-oxide material. The maximum idler efficiencies of 6% with HGS-1 and 2.1% with HGS-2 correspond to quantum conversion efficiency of 23% and 12.5%, respectively, lower than the measured pump depletion, due to intracavity idler losses not taken into account when calculating them from the idler output energy.

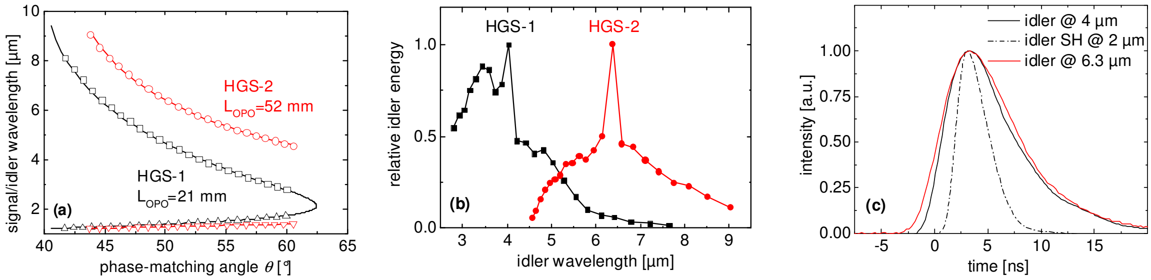

Fig. 2. Experimental signal and idler OPO wavelengths (symbols) vs. internal phase-matching angle θ compared with calculations (lines) (a), relative idler energy vs. idler wavelength (b), and temporal pulse profiles of the idler at 4 µm, its SH at 2 µm, and of the idler at 6.3 µm (c).

Figure 2a shows a comparison of the experimental OPO tuning with calculations based on the Sellmeier equations for HGS that gave the best agreement [5]. The idler tuning range extends from <3 to >8 µm with HGS-1 and from ~4.5 to >9 µm with HGS-2. The "anomalous" peak at normal incidence for both crystals is due to enhanced feedback, especially for the idler wave. The OPO linewidth, measured at the signal wavelength with a 1-mm-thick Ag-coated CaF_2 Fabry-Perot etalon, was 1.9 cm^{-1} or 0.4 nm at 1446 nm for HGS-1 and ~2.3 cm^{-1} or ~0.4 nm at 1280 nm for HGS-2. The temporal characteristics of the HGS OPOs were studied with fast photodiodes and 0.5 GHz oscilloscope. Taking into account the finite time response of the (HgCdZn)Te detector used for the idler and its second-harmonic (SH, generated in a 3-mm thick type-I GaSe crystal and detected by InGaAs photodiode) pulse profile one arrives at an actual idler pulse duration of ~5 ns near 4 µm, and <7 ns at 6.3 µm, both shorter than the pump pulses. The M^2 beam quality factor of the HGS OPOs, measured by the knife-edge method, amounted to M^2~180-190 in the two planes with HGS-1, with HGS-2 it was ~6-times better, M^2~30, due to the longer cavity. The relatively high M^2 values can be attributed to the large pump diameter, short pump pulse duration and operation far above threshold. It is expected that this parameter could be substantially improved implementing the RISTRA cavity OPO concept [6] and such experiments are ongoing.

We conclude that HGS holds great potential for coverage of the mid-IR range up to ~12 µm with OPOs pumped near 1 µm (powerful Nd- or Yb-based pulsed laser systems). Output energies on the 10 mJ level (i.e. >1 W) and >5 mJ above 5 µm can be expected using commercially available pump sources at 1064 nm. Reduction of OPO threshold using longer HGS samples will help avoid optical damage, increase the repetition rate for higher average power, and operate at least three times above threshold also at longer idler wavelengths. The same can be achieved using flat top pump beam shaping and/or RISTRA cavity design, which has proved to be the best solution for reaching highest idler pulse energies in this mid-IR spectral range from similar OPOs pumped near 2 µm [6].

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