Widely Tunable in the Mid-IR $BaGa_4Se_7$ Optical Parametric Oscillator Pumped at 1064 nm

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Abstract: A $BaGa_4Se_7$ optical parametric oscillator shows extremely wide idler tunability (2.7-17 µm) under 1.064-µm pumping. The ~10-ns pulses at ~7.2 µm have an energy of 3.7 mJ corresponding to a quantum conversion efficiency of 40%.
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1. Introduction

Only few non-oxide nonlinear optical crystals can be used for frequency down-conversion of high-power solid-state laser systems operating near 1 µm (e.g. Nd:YAG at 1.064 µm) to the mid-IR (3-30 µm) and in particular beyond ~5 µm, the upper wavelength cut-off limit of oxide based materials [1]. From the non-oxide crystals that are free of two-photon absorption (TPA), i.e. exhibit a bandgap corresponding to <0.532 µm, only very few have been applied in short (ns) pulse pumped Optical Parametric Oscillators (OPOs) at 1.064 µm. The chalcopyrite type $AgGaS_2$ (AGS), the only commercially available such crystal, has generated so far the longest idler wavelengths for 1-µm pumped OPOs, 11.3 µm [2], while the highest output energies beyond 5 µm, 3 mJ at 6.3 µm, were achieved with the related defect chalcopyrite $HgGa_2S_4$ (HGS) [1] which is extremely difficult to grow in large sizes.

Recently, we added a newly developed chalcogenide crystal, $BaGa_4S_7$ (BGS) with orthorhombic *mm*2 structure to this short list of non-centrosymmetric nonlinear crystals of this kind that can be pumped at 1.064 µm [3]. It showed a tunability range from ~5.5 to ~7.3 µm with a maximum energy of ~0.5 mJ at ~6.2 µm. Its selenide counterpart $BaGa_4Se_7$ (BGSe) is expected to exhibit much higher nonlinearity [4]. BGSe is also biaxial but monoclinic, i.e. it offers much more phase-matching options [4,5]. It shows a transparency extending from 0.776 to 14.72 µm at the 0.3 cm^{-1} absorption level, however the bandgap value, 2.64 eV, corresponds to 0.469 µm, i.e. no TPA is expected at 1.064 µm. Figure 1(a) shows a typical transmission spectrum of one of the BGSe samples used in the present work. Larger bandgap normally leads to higher damage threshold. With 14 ns pulses at 100 Hz we estimated a damage threshold of 1.4 J/cm^2 leading to a peak on-axis intensity limit of 100 MW/cm^2.

A BGSe OPO has already been reported but pumped at 2.091 µm [6] where the damage threshold is higher. Nevertheless, this typical 3-5 µm OPO essentially did not show tunability above the ~5 µm oxide crystal limit. Here we demonstrate, for the first time to our knowledge, optical parametric oscillation in the mid-IR based on BGSe pumped at 1.064 µm. We report record long idler wavelengths (17 µm) for BGSe, achieving the highest conversion efficiency and the highest output idler energy above 5 µm (oxide materials limit) for any OPO pumped at 1.064 µm.

2. Experimental set-up and results

From preliminary (unpublished) results on second-harmonic generation (SHG) and some partially resolved components of the 2nd order susceptibility tensor reported in [7] we decided to investigate two active elements made of BGSe. BGSe-I was a sample cut at $\theta = 46°$ for ee-o positive type-I interaction in the *x*-*z* plane. Its dimensions were 10.33 (along *y*-axis) × 11.95 × 14.57 (length) mm^3. The second sample BGSe-II was cut at $\theta = 33.5°$ for oe-o positive type-II interaction in the *y*-*z* plane. This sample was slightly shorter, with dimensions of 9.22 (along *x*-axis) × 11.32 × 13.56 (length) mm^3. Both samples were AR-coated for increased transmission at the signal wave which resulted in a good transmission also for the pump at 1.064 µm, see Fig. 1(a). According to the monoclinic class *m* symmetry, the corresponding expressions for the effective nonlinearity of BGSe read:

$$d_{eff}(x-z) = d_{16}\cos^2\theta + d_{23}\sin^2\theta\,, \qquad (1)$$

in the *x*-*z* plane and

$$d_{eff}(y-z) = \pm d_{16}\cos\theta - d_{15}\sin\theta\,, \qquad (2)$$

in the *y-z* plane. The tensor components are defined in the orthogonal dielectric frame *xyz* ($n_x < n_y < n_z$) and under Kleinman symmetry $d_{16} = d_{21}$, $d_{23} = d_{34}$, and $d_{15} = d_{31}$. For BGSe the monoclinic *b*-axis coincides with the dielectric *x*-axis [5]. Unfortunately, the information on the components of the 2nd order susceptibility tensor is rather limited [7,8]. From the evaluated non-diagonal nonlinear coefficients, d_{23} is the largest and d_{15} is negligible compared to it. The lack of information on d_{16} motivated us to study the two cases.

The singly resonant OPO was set-up with a standard linear cavity consisting of a flat input-output coupler (IOC) and a flat Au total rear reflector which ensures recycling of the pump and a double pass for the non-resonant idler prior to its extraction through the IOC, the same as the one shown in [3]. Pumping via a 45° ZnSe bending mirror which was highly transmitting for the signal and idler ensured separation of the input and output waves. Since the IOC was highly transmitting for the pump and idler and highly reflecting for the signal, the output consisted basically of the idler, which was characterized behind the bending pump mirror.

The pump source was a 100 Hz diode-pumped Nd:YAG master oscillator power amplifier system providing pulses of 8 ns duration with an energy of up to 250 mJ [3]. The output beam was passed through an attenuator (half-wave plate and polarizer) and a vacuum diamond pinhole and then down collimated by a lens telescope to a Gaussian diameter of 5 mm in the position of the OPO. The spectral bandwidth was ~30 GHz (1 cm^{-1}) and M^2 ~ 2.

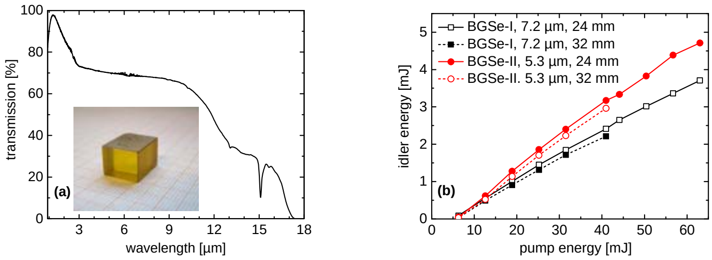

Fig. 1. (a) Unpolarized transmission of the AR-coated sample BGSe-II (shown as inset) and (b) input-output characteristics of the BGSe OPO at normal incidence for a cavity length of 24 and 32 mm.

Focusing on the energetic performance of BGSe in the OPO we reduced the repetition rate of the pump laser by means of an external shutter to 10 Hz. The main reason for this was the damage limit we observed for all the available metallic total reflectors when operating this system at 100 Hz [3]. Figure 1(b) shows the input-output OPO characteristics at normal incidence for a short (24 mm) and slightly longer (32 mm) cavity. The antireflection (AR)-coated CaF_2 IOC shows no substrate absorption at the actual idler wavelengths and the transmission is ~85% (6.3 µm). Its reflectivity measured at the signal wavelength (1.28 µm) is 73%. The pump energy given is the one incident on the BGSe crystals while the idler energy is the one behind the IOC. The results with the 24-mm cavity in Fig. 1(b) present the highest energy achieved in the mid-IR above 5 µm with a 1-µm pumped OPO [1]: note that BGSe-I generated 3.7 mJ at a wavelength of 7.2 µm, longer than the one reported for HGS (6.3 µm) in [1], i.e. at decreasing parametric gain. With BGSe-II, a maximum energy of 4.7 mJ is obtained at 5.3 µm. This energy is much higher than the maximum output achieved at the longest idler wavelength of 5.19 µm (estimated to be < 1.4 mJ from the total output presented in Fig. 2 in [6]) reported for the 2.091 µm pumped BGSe OPO.

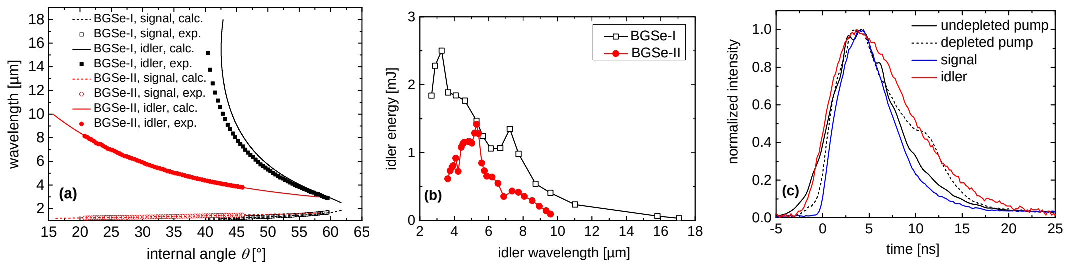

Fig. 2. (a) Angle tuning of the OPO with BGSe-I and BGSe-II versus internal phase-matching angle: symbols (experimental data) and curves (calculated with Sellmeier expressions from [9]), (b) Output idler energy versus wavelength for BGSe-I and BGSe-II. The cavity length is 32 mm and the pump energy 27 mJ, and (c) Simultaneously measured temporal pulse shapes of the undepleted and depleted pump, signal and idler for BGSe-II at normal incidence at a pump energy of 31.5 mJ.

The threshold is around 6 mJ for both BGSe crystals. The maximum pump level applied (63 mJ) corresponds to an axial fluence of 0.64 J/cm^2 or a peak on-axis intensity of 80 MW/cm^2, still below the damage threshold. However, pump depletion also helps to avoid damage to the BGSe samples in this highly efficient OPO. Assuming same number of signal photons generated, the pump depletion is 40% for BGSe-I and 37% for BGSe-II, almost 3 times higher than the total conversion efficiency reported in [6] near degeneracy. At maximum pump level, the corresponding pump to idler energy conversion efficiencies are 5.9% and 7.5% and the slope efficiencies: 6.5% and 8.3%. Comparing to the HGS OPO [1], the pump depletion achieved with BGSe-I is >3 times higher, the pump to idler efficiency almost 3 times higher and the slope efficiency – 2 times higher. Thus the quality of the present BGSe samples seems to be excellent. Note that for HGS, at the maximum pump levels applied [1] which were very similar to the present work in terms of fluence and intensity, formation of scattering centers in the bulk was observed.

Tuning was studied by tilting the crystals in a lengthened cavity (32 mm). The AR-coated ZnSe IOC employed for these measurements shows high transmission for the idler in a narrow spectral range for which it was optimized (95% at 6.3 µm). The experimental data for BGSe-II in Fig. 2(a) are in excellent agreement with calculations based on the Sellmeier equations presented in [9]. For BGSe-I, deviations exceeding 1° are seen, especially above 11 µm where the Sellmeier equations in [9] were not fitted.

The idler tuning range in Fig. 2(b) extends from 2.7 to 17 µm for BGSe-I and from 3.6 to 9.6 µm for BGSe-II. This result presents the longest wavelength ever achieved with a 1-µm pumped OPO [1,2]. BGSe delivered much wider OPO tunability compared to its sulfur counterpart BGS [3]. Typical enhancement is observed for both crystals at normal incidence (7.2 and 5.3 µm, respectively) where additional feedback is provided by the Fresnel reflections. The one conclusion that can be drawn from the performance of BGSe-I and Eq. (1) is that the nonlinear coefficients d_{16} and d_{23} must have the same sign, contrary to the theoretical predictions in [4]. On the other hand, the performance of BGSe-II with Eq. (2) leads to the conclusion that d_{16} is substantially larger than d_{15}, i.e. it must be comparable in magnitude to d_{23}.

The OPO linewidth for the signal wave was measured at normal incidence using a 1-mm-thick Ag-coated CaF_2 Fabry-Perot etalon: it was ~40 GHz both for BGSe-I and BGSe-II. Convolution of this value with the spectral extent of the pump gives a spectral FWHM of 9 nm at 7.2 µm (BGSe-I) and 5 nm at 5.3 µm (BGSe-II). Figure 2(c) shows the temporal characteristics of the pulses measured for BGSe-II with a fast InGaAs photodiode (pump and signal) or a (HgCdZn)Te detector with a time constant of <2 ns (idler). As a result of depletion and back conversion the pump pulse is reshaped and lengthened. The ~10 ns FWHM of the idler at 5.3 µm can be considered as an upper limit at high conversion efficiency due to the limited temporal resolution. Very similar results were obtained with BGSe-I at 7.2 µm. Thus, the output pulse durations are rather close to that of the pump which means short build-up time resulting in high conversion efficiency. The idler beam spatial profiles were recorded by a SpiriconTMPyrocam III camera with $LiTaO_3$ pyroelectric detector (active area: 12.4×12.4 mm^2, element size: 0.1×0.1 mm^2). At a pump level of 31.5 mJ and normal incidence, the M^2 values obtained for the idler were between 40 and 60 in the two planes, both for BGSe-I and BGSe-II. Other factors that contribute to these high values for the non-resonated wave, besides the high conversion efficiency far above threshold, are the large Fresnel number and the short pump pulse duration.

In conclusion, the newly developed monoclinic BGSe showed excellent optical quality and performance in a 1.064 µm pumped OPO. It generated the highest pulse energy for a non-oxide crystal above the 5 µm limit of oxide materials. Unprecedented tuning from 2.7 to 17 µm could be achieved with a single crystal cut. Pump to idler conversion efficiencies exceed previously reported values by a factor of ~3 and the pump depletion reached 40%.

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