Widely Tunable (2.2 – 10.4 μm) $BaGa_4Se_7$ Optical Parametric Oscillator Pumped by a Q-switched Nd:$YLiF_4$ Laser

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Abstract: We report on the first $BaGa_4Se_7$ nanosecond optical parametric oscillator pumped by Q-switched Nd:$YLiF_4$ laser at 1053nm. Mid-infrared idler wave tuning from 2.2 μm to 10.4 μm is demonstrated with an angle-tuned type-I (o-ee) *y*-cut sample.
OCIS codes: (190.4400) Nonlinear optics, parametric processes; (190.4400) Nonlinear optics, materials; (140.3600) Lasers, tunable.

1. Introduction

Widely tunable optical parametric oscillators are attractive laser sources for mid-infrared (MIR) nonlinear optics applications, because of the scarcity of direct-emitting conventional laser sources in this spectral region (2 - 20 μm). For instance the 8 – 14μm range corresponding to one of the atmospheric transparency window is interesting for LIDAR or military applications (counter-measure systems). In military applications the atmospherically transparent band II (3-5 μm) and band III (8-14 μm) also motivate the search for performant mid-IR nonlinear compounds. On fundamental physics ground, the development of intense pulsed MIR laser sources opens new ways to study the interaction of high-intensity laser radiation with, taking profit of the quadratic wavelength dependence of the pondemorotive electron energy ($U_g \sim I\lambda^2$) which relaxes the laser intensity requirement [1].

The continuous coverage of the MIR spectrum from 5 to 20μm requires thus the continuous synthesis of novel nonlinear birefringent ternary chalcogenide compounds, combining both wide energy bandgap and extended MIR transmission. Recently two new promising ternary chalcogenide compounds transparency have been successfully synthesized in large size using the Bridgman-Stockbarger technique: $BaGa_4S_7$ (BGS) and $BaGa_4Se_7$ (BGSe) [2-4]. Both sulfide and selenide compounds exhibit wide bandgap energy (E_g = 3.54 eV with λ_g = 0.350mm for BGS; E_g = 2.64 eV with λ_g = 0.469μm for BGSe) and extended MIR transparency (up to 12 μm for BGS and from 0.47 to 18μm for BGSe at 0% transparency level). Owing to their large bandgap, these compounds exhibit one of the largest damage threshold among MIR nonlinear compounds. The first BGS nanosecond OPO pumped by a Nd:YAG laser at 1064 nm was demonstrated by Tyazhev *et al* [5], in which the idler wave could be tuned from 5 to 7.3 μm. While BGS crystallizes in the *mm*2 orthorhombic point group, BGSe crystallizes in the monoclinic *m* point group (with the dielectric axes *x* and *z* aligned respectively with the *b* and *c* crystallographic axes [3,4]), offering thus an extended phase-matching capability than BGS. The first BGSe nanosecond OPO ever reported was pumped by a Q-switched Ho:YAG laser at 2.09 μm and used a *y*-cut crystal ($\theta = 40.8^o$, $\varphi = 0^o$) for type-I (o-ee) phase-matching in the *xz* plane [6]. Recently a second 1064-nm pumped BGSe OPO was reported by Kostyukova *et al*, demonstrating an unprecedented output performance in terms of both idler MIR tunability (2.7 – 17 μm, i.e. over its full MIR transparency range) and output pulse energy (3.7 mJ at 10 Hz at 7.2 μm) [7].

In this communication, we report on a $BaGa_4Se_7$ nanosecond OPO pumped for the first time by a Q-switched Nd:$YLiF_4$ (Nd:YLF) at 1053 nm, the shortest OPO pump wavelength used for this crystal. Contrary to [7], the pump laser has not only a slightly shorter wavelength but also much lower pulse energy (maximum pulse energy 1.3 mJ). Low threshold of oscillation ($E_{p,th}$ = 0.25 mJ) was achieved by reducing the pump beam diameter to $2w \sim$ 0.5 mm, thus the reported performance corresponds to the operation of the OPO far below parametric gain saturation (and damage threshold). The MIR idler wave tunability extends from 2.2 μm to 10.4 μm, greater than for a mercury thiogallate (HGS) OPO pumped by the same Nd:YLF laser and employing an identical crystal length [8]. We report on surface damage threshold measurements of BGSe at 1053nm, confirming the high damage threshold (>2 J/cm^2 for 10-ns pulse and f = 100 Hz repetition rate) reported in [3] for similar experimental conditions. Additionally, with our phase-matched OPO data, we could infer the accuracy of three recently published dispersion and thermo-optic dispersion relations of $BaGa_4Se_7$.

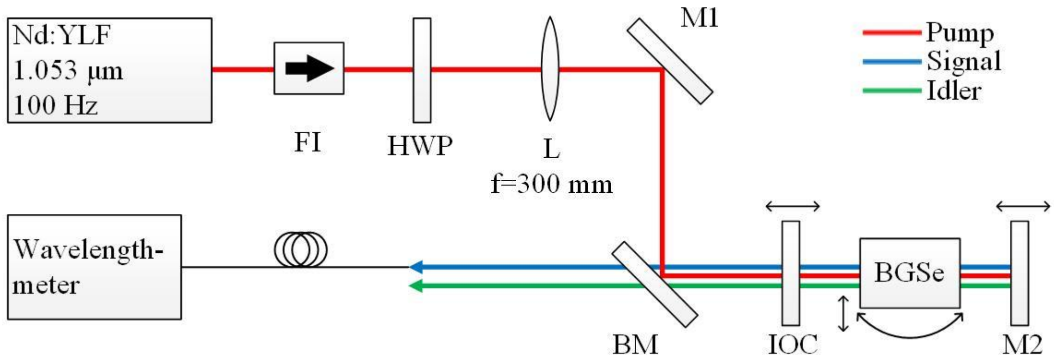

Fig. 1. Experimental $BaGa4Se_7$ OPO layout. HWP: Half-wave plate; BM: dichroic bending mirror (HR @ 1-1.26 μm, HT @ 1.3 – 10 μm); IOC: input-output ZnSe coupler (T = 93% @ 1053 nm, partial HR @ 1.17 – 2.1 μm); M2: gold mirror. The *xz* plane of BGSe is within the plane of the drawing. The sample (dimensions 3 × 7 × 11.8 mm^3) was cut for type-I (o-ee) phase-matching ($\theta = 45^o$, $\varphi=0^o$) and was single-layer AR coated at ~ 1.5 μm.

2. Experimental setup

The experimental layout of the BGSe OPO is sketched in figure 3. The pump source is a diode-pumped Q-switched Nd :YLF laser (Laser-compact Group model TECH-1053-N). The pump wavelength is 1.053 μm and the pulse duration is13 ns, with a pulse repletion rate set to 100 Hz. The specified bandwidth of the laser is ~ 1 cm^{-1}, within the pump acceptance bandwidth of the type-I phase-matching process. The maximum laser pulse energy is 1.3 mJ and the measured beam quality factors are $M^2 \approx 1.2$. The OPO cavity, configured as a double-pass pump, signal-resonant oscillator (SRO), is formed by the input-output ZnSe coupler (IOC) and a gold (Au) mirror M2.

The cavity length was kept as short as possible ($L_{cav} \geq 18$ mm) to increase the number of signal-wave roundtrips within the cavity, except for idler wavelength tuning purpose for which the cavity is lengthened to accommodate angular tuning. The BGSe sample was hold inside a copper mount that is temperature-regulated at T = 40^o C by means of a thermo-electric Peltier element. For wavelength tuning purpose, the BGSe mount is positioned on a motorized high-precision rotation mount (STANDA model 8-MR-190-2). The signal wavelength was measured by a commercial wavemeter (ANGSTROM model *HighFinesse* WS6 IR1) and the idler wavelength was retrieved from energy conservation. The idler wave behind BM is filtered from the leaking signal wave by two additional dichroic mirrors (not shown), but the signal wave could not be totally blocked below 2.6 μm, rendering the idler power measurement inaccurate for the shorter part of the idler branch down to the degeneracy point at $2\lambda_p$.

3. Experimental results

Figure 2a shows the BGSe OPO idler pulse energy at λ_i = 8.07 μm (corresponding to sample at normal incidence and a short cavity length L_{cav} = 18 mm) as the pump pulse energy is increased, yielding a pump threshold as low as $E_{p,th}$ = 250 μJ. The data could be excellently fitted with a straight line, showing no sign of saturation as expected since the BGSe OPO is pumped even farther from parametric gain saturation. Comparing with the performance of a ns OPO pumped with the same Nd:YLF laser and employing a similar length mercury thiogallate (HGS cut for type-II eoe at $\theta = 47^o$, $\varphi = 0^o$, with a 4× larger effective nonlinear coefficient), and which yielded 2 μJ of idler at λ_i = 7.5 μm (normal incidence) [8].

Figure 2b displays the measured idler energy measured after IOC mirror as a function of wavelength. For reasons explained in the previous section, we did not plot the data below λ_i = 2.6 μm down to 2.2 μm because of imperfect separation of the idler energy from the leaking signal energy. The MIR coverage (2.2 – 10.5 μm) obtained with a single BGSe crystal exceeds the one (5.6 – 10.8 μm) obtained from the previously quoted HGS OPO, that needed two different samples to cover that idler range [8]. This broad coverage with a single sample highlights the broader phase-matching capability of the monoclinic $BaGa_4S_7$ compared with the uniaxial defect chalcopyrite $HgGa_2S_4$. With larger available pulse energy, this MIR range could have been extended up to 17 μm as done in [7]. We have investigated damage threshold at 1053nm by focusing the laser beam down to a 2ω = 200 μm diameter, using several uncoated test plate cut from the same ingot as the present OPO sample. The highest damage threshold was measured to the (2.04 ±0.39) J/cm^2 for f =100 Hz repetition rate. At f = 150 Hz, it is almost unchanged to (2.02 ±0.31) J/cm^2 and drops to (1.81 ± 0.25) J/cm^2 at f = 150 Hz.

The phase-matching angles associated with the MIR tuning curve shown in figure 2b are plotted in figure 3. As seen the OPO idler wave could be tuned from nearly the degeneracy point to 10.5 μm.

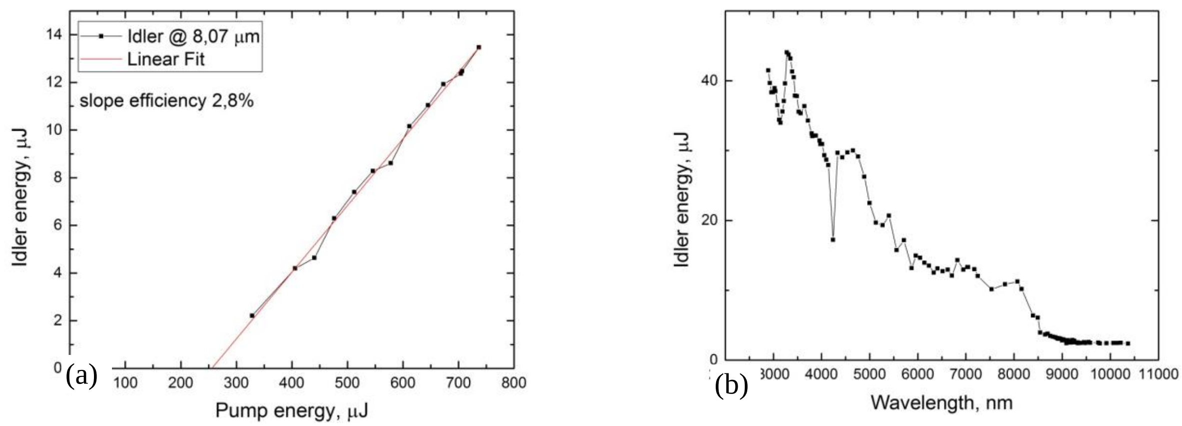

Fig. 2. (a) Measured idler energy at ~8.1 μm versus pump pulse energy measured in front of the OPO cavity (after BM); (b) MIR pulse energy versus wavelength. The data between 2.2 and 2.6 mm were discarded because the two dichroic filters could not efficiently filter the leaking signal wave.

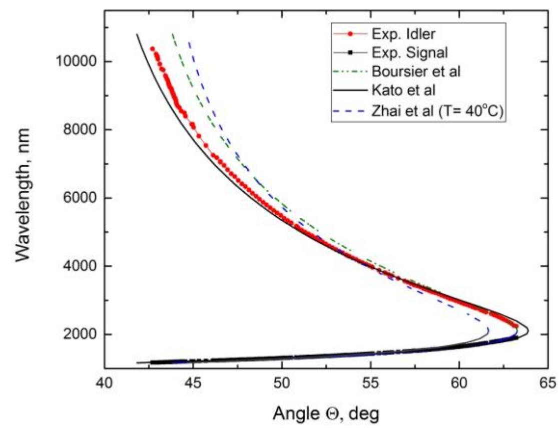

Fig. 3. Nd:YLF pumped (λ_p = 1053 nm) type-I (o-ee) $BaGa_4S_7$ OPO phase-matching curve.

Very recently new dispersion formulae (Sellmeier equations) for BGSe have been proposed by three different groups [9-11]. We found out that the most accurate ones, constructed from various phase-matching datasets spanning the visible to MIR wavelength up to 11 mm, are given by Ref. [11].

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