





High Power and Efficient 4.43 μm BaGa₄Se₇ Optical Parametric Oscillator Pumped at 1064 nm

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Abstract: A high power and efficiency mid-infrared optical parametric oscillator based on a $BaGa_4Se_7$ crystal is demonstrated in this paper. It was pumped by a 500 Hz Q-switched Nd:YAG laser at room temperature. Without cooling, up to 0.76 W output power at 4.43 µm was generated with respect to the incident pump power (1064 nm) of 5.52 W, corresponding to an optical-to-optical conversion efficiency of 13.7%. The corresponding slope efficiency was as high as 18.7%. The pulse width of the signal wave was 5.2 ns at the pump pulse of 13.7 ns. To the best of our knowledge, this is to date the highest output power achieved at 4–5 µm from a 1064 nm pumped BGSe OPO laser. Considering that no additional cooling system was applied, this work provides a good solution for a high-efficient, compact or even portable mid-infrared solid-state laser device.

Keywords: mid-infrared lasers; nonlinear optics materials; parametric oscillators and amplifiers; no cooling



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1. Introduction

Mid-infrared (MIR) 4–5 µm laser sources have attracted enormous attention over the last decades. They have great application prospects in remote sensing, environmental monitoring, medical treatment, and atmospheric detection [1–3]. Optical parametric oscillators (OPOs) are an effective approach to obtain a $4-5 \mu m$ laser due to the distinctive advantage of being highly compact and efficient. The nonlinear optical (NLO) frequency conversion crystals with wide range transparency, large nonlinear coefficient, and high laser damage threshold are considered as the key elements to generate the MIR laser radiation. Currently, a large number of NLO crystals (oxide and non-oxide) have been investigated for the MIR region [4]. However, due to the two- or multi-phonon absorption phenomena, oxide crystals are difficult to be used to generate the MIR radiation above 4 µm. Some non-oxide crystals, with wider transparency spectrum, are more suitable for wavelength radiation longer than 4 µm. BaGa₄Se₇ (BGSe) crystal is a promising NLO crystal for MIR laser generation. It has a wide bandgap (2.64 eV), large NLO coefficient (24.3 pm/V), large damage threshold (557 MW/cm²), and wide transparency spectrum (0.47–18 μ m) [5–8]. The BGSe has been extensively used in OPO operation at MIR region pumped by economical ~1 µm laser sources, such as pulse Nd:YAG lasers [9–14].

To date, some high energy and high peak power MIR lasers have been obtained from the nanosecond 1064 nm pumped BGSe OPOs. In 2016 [9], a 1064 nm pumped tunable MIR (2.7–17 μ m) BGSe OPO was reported for the first time, with an ~10 ns output pulse at ~7.2 μ m and energy of 3.7 mJ at 10 Hz. In 2017 [11], Xu et al. presented a high pulse energy MIR BGSe OPO. They achieved the energy of 2.56 mJ (10 Hz) at 4.11 μ m, with the peak power of 256 kW. Recently [12], our group has reported a more compact and efficient BGSe OPO cavity. A 4.25 μ m laser pulse with the energy of 1.03 mJ (10 Hz) was

obtained under 13.5 mJ pump energy, corresponding to a high conversion efficiency of 7.6% and a slope efficiency of 12%. Up to now, most of the 1064 nm pumped BGSe OPOs operating at a low frequency rate (10 Hz/100 Hz) were less than 100 mW. Instead, longer wavelength pump sources, such as 2.01 μ m and 2.79 μ m, are promising to achieve MIR lasers with higher power and a high frequency rate (500 Hz/1 kHz) due to the higher laser damage threshold and higher quantum efficiencies [15–17]. In 2016, a BGSe OPO was pumped by a 2090.6 nm Q-switched Ho:YAG laser for the first time [15]. It generated the maximum output power of 1.55 W at 3.96 μ m and 4.39 μ m with 500 Hz. At 1 kHz, the output power was further increased to 1.68 W at 4.5 μ m, with a conversion efficiency of 18.3% and a slope efficiency of 30.0% [16]. However, compared with the widely used 1 μ m pump sources, the generation of high power 2 μ m pump sources is much more complicated and expensive. Therefore, if the high power and highly efficient BGSe OPOs could be generated with the more economical 1 μ m laser as a pump source, it is promising to achieve the commercialization of such MIR lasers.

In this paper, we demonstrate a highly efficient and compact MIR BGSe OPO system. It is operated without any cooling. A high-quality BGSe NLO crystal and a 500 Hz Q-switch Nd:YAG laser are employed as the gain medium and pump source, respectively. Up to 0.76 W average output power at 4.43 μ m is successfully obtained from the BGSe OPO. To the best of our knowledge, this is the highest reported output power achieved to date from a 1064 nm pumped BGSe OPO laser at above 4 μ m radiation.

2. Materials and Experimental Setup

The growth of BGSe crystals was reported in 2010 for the first time [5]. Recently, to further improve the optical properties of the crystals, a refined two-temperature-zone growth technique was employed for the polycrystalline material fabrication [18]. Using two-temperature-zone vapor transfer, high-quality and large-size BGSe single crystals were grown successfully, with a high transmittance from 0.47 μ m to 18 μ m and a calculated absorption coefficient of around 0.04 cm⁻¹. Additionally, then, via two zone thermal annealing method, a better annealing effect was obtained than before. As shown in Figure 1, the newly grown 15 mm long BGSe crystal, with optimized polishing and coating, showed enhanced optical properties, such as lower absorption and lower laser damage threshold, which was adopted in our experiment to gain a higher average power.



Figure 1. Photograph of the BGSe crystal: (a) side view and (b) end view.

The experimental configuration of the BGSe OPO is shown in Figure 2. The pump source was a homemade 1064 nm pulse laser in our laboratory. It is an diode side-pumped electro-optic Q-switching Nd:YAG laser at 500 Hz. The pulse width was 13.7 ns at the maximum output pulse energy of 11 mJ (corresponding to an average power of 5.5 W). The pump pulse was firstly transmitted through an isolator to avoid possible damage to the Nd:YAG cavity caused by the reflected 1064 nm laser beam from the OPO cavity. In order to obtain a fixed o-polarized light, a half-wave plate (HWF) and a polarized beam splitter (PBS) were inserted between the isolator and OPO cavity. By turning the HWF's angle, the inpouring pump power can be adjusted to the required value. The BGSe OPO cavity was composed of three parts: the BGSe NLO crystal and two flat mirrors. The BGSe crystal was cut into a size of 15 mm in length and 6 mm \times 6 mm in cross-sectional dimension.

To achieve o-ee Type-I phase matching at 4.3μ m, the *x*–*z* plane was cut at $\theta = 53.2^{\circ}$ and $\varphi = 0^{\circ}$. Both end surfaces of the crystal were laser-grade polished and anti-reflection (AR) coated at the pump, signal and idler laser wavelengths. The crystal was mounted in an aluminum clamper without cooling. The input mirror M1 was AR coated at 1064 nm for one surface, and highly transmission (HT, T > 99%) coated at 1064 nm and highly reflection (HR, R > 98%) coated at 3.8–4.7 µm for the other surface. The output coupler M2 with transmission of 40% at 3.8–4.7 µm was utilized to investigate the output performances of the BGSe crystal laser. It was HR (R > 99%) coated at 1064 nm for the double-pass-pump OPO to reduce the threshold energy and improve the output power. The cavity length was adjusted to 50 mm. The pump beam was transmitted to the BGSe crystal with a pump spot diameter of 3.6 mm. Moreover, a dichroic mirror M3 was 45°HR coated for idler wavelength region and AR coated for signal wavelength region, to separate the output idler and residual signal laser beams.



Figure 2. Schematic diagram of the experimental setup.

3. Results and Discussions

The output spectrum recorded was monitored by an optical spectrum analyzer (Ocean Optics, NIR Qust-256, 870–2500 nm) outside the OPO cavity. Merely 1% energy of pump beam could be transmitted through the output coupler M2, i.e., the residual pump light was reserved in the output laser beam from the OPO cavity. Both the pump and signal waves were measured at the same position through a coupling fiber. Figure 3 shows the typical output spectral recorded outside the OPO cavity (behind the mirror M2). Residual pump laser of $\lambda_p = 1064$ nm was detected outside the cavity. In addition, obvious signal peak was observed at $\lambda_s = 1400$ nm. Based on the OPO moment conservation condition $(1/\lambda_p = 1/\lambda_s + 1/\lambda_i)$, the idler wavelength λ_i was calculated to be 4.43 µm.



Figure 3. The typical spectral recorded of the pump and signal wave.

Furthermore, the measured transmissivity at 1400 nm signal wave for M1 and M2 were about 40% and 60%, respectively. The mid-infrared OPO was an idler wave single resonant oscillator (SRO). Both the double-pass pump and SRO can effectively reduce the OPO pump threshold and increase the idler output power [19]. The output power of idler light was measured by a power meter (Newport, 919-003-10, 0.19 μ m–11 μ m, with maximum range of 3 W). The employed maximum pump power was 5.52 W, along with 11.08 mJ pulse energy and 13.7 ns pulse width. The output power of the 4.43 μ m idler light versus the input 1064 nm pump power is shown in Figure 4. As depicted in the curve, the output

power increased linearly with the increase in pump power, while no power saturation was detected. The threshold pump power of OPO was 1.72 W, corresponding to the pump energy of 3.4 mJ and peak intensity of 2.5 MW/cm^2 . The maximum output power of 0.76 W was obtained at 4.43 µm under the incident pump power of 5.52 W, corresponding to an optical-to-optical conversion efficiency of 13.7% and a slope efficiency of 18.7%. To the best of our knowledge, this is the highest output power above 4 µm so far achieved on the 1064 nm pumped BGSe OPO.



Figure 4. Output power of the idler light versus the input pump power.

However, when the multi-watts high power pump light was employed to double pass through the BGSe crystal in our experiment, the heating effect of absorbed light should cause a temperature increase inside the crystal. The temperature of the side surface for the BGSe crystal was measured by an infrared camera (FLIR T620). As shown in Figure 5, the temperatures increased with the increase in pump power, and reached to 39 $^{\circ}$ C at the maximum pump power of 5.52 W. The thermal conductivity coefficients along the three crystallographic axes of the BGSe crystal were 0.74, 0.64, and 0.56 W·m⁻¹·K⁻¹ [6]. Due to the relatively low thermal conductivity and the non-uniform heat conduction, there is a higher temperature distribution in crystal's center region, and the thermal lens effect was easily induced during the high power operation. It known that the relationship coefficient between thermal refractive index and temperature was about 3.2 nm/°C [20], and the calculated phase matching curves of BGSe are varied at different temperatures. This is the reason that, in the current work, the output 4.43 µm idler wavelength is longer than the value (4.3 µm) calculated from the phase matching curves at room temperature. In addition, the inhomogeneous temperature distribution should reduce the conversion efficiency of the OPO. Additionally, it is expected that further improvements will be implemented by employing active cooling for the BGSe crystal, which should obtain watt-level and multi-watts level MIR lasers.



Figure 5. The surface temperature of the BGSe crystal versus the pump power.

The beam profile of the signal light was measured by a high-quality CCD camera (Thorlabs, BP209-IR, 900–1700 nm) behind the dichroic mirror M3. When the pump power was increased to 5.52 W, the output idler laser power reached the maximum of 0.76 W. The corresponding beam profile of the output signal light is shown in Figure 6. It has a near Gauss mode profile, with the Gaussian fit diameters of 5.66 mm in x-direction and 5.62 mm in ydirection. Figure 7 illustrates the temporal profiles of the pump light and signal output light, which were measured by a 2 GHz digital oscilloscope, a photo-detector (rise time < 175 ps, 170–1100 nm) and the other fast photo-detector (rise time < 35 ps, 350–1700 nm). Under the pump power of 5.52 W, the duration of the pump pulse (Figure 7a) was 13.7 ns, while that of the output signal pulse (Figure 7b) achieved merely 5.2 ns. According to the OPO energy conservation condition, the signal and idler photons are generated in pairs, resulting in the same temporal property between the signal pulse and idler pulse [21,22]. Therefore, the signal wave monitored from the OPO cavity should have well indicated the temporal behavior of the idler wave.



Figure 6. Beam profile of the output signal light.

(a)	(b)	
13 7 ns		
		5.2 ns
	alternation and an approximation	
20 ns/div		20 ns/div

Figure 7. Temporal profiles of the pump light (a) and output signal light (b).

4. Conclusions

In conclusion, with a high-quality BGSe crystal as the gain medium and a 500 Hz Q-switched Nd:YAG laser as the pump source, a high power and efficient laser operation at 4.43 μ m was obtained at room temperature. To further improve the compactness of the system, an additional cooling system was not applied. The maximum output average power of 0.76 W was achieved at a pump power of 5.52 W. This is the highest output power at above 4 μ m radiation reported so far from a 1064 nm pumped BGSe OPO laser, to the best of our knowledge. The optical-to-optical conversion efficiency from 1064 nm to 4.43 μ m was 13.7%, with the slope efficiency of 18.7%. The pulse width of the signal wave was measured to be 5.2 ns at a pump pulse of 13.7 ns. Since additional cooling was not applied, such devices was highly compact and could be readily applied in various occasions, such as laser raging, navigation, environmental monitoring and laser scalpel.

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