Narrow-linewidth BaGa₄Se₇ optical parametric oscillator

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The linewidth of the BaGa₄Se₇ (BGSe) optical parametric oscillator (OPO) was narrowed for the first time, to the best of our knowledge, by inserting a Fabry-Perot (FP) etalon into an L-shaped cavity. When a 15 mm long BGSe (56.3°, 0°) was pumped by a 1064 nm laser, the peak wavelength was ~3529 nm, and the linewidth was 4.53 nm [3.64 cm⁻¹] under type I phase matching. After inserting a 350 μm thick FP etalon, the linewidth was decreased to 1.27–2.05 nm. When the tilt angle of the etalon was 2.34°, the linewidth was 2.05 nm (1.65 cm⁻¹), and the peak wavelength was still ~3529 nm. When the tilt angle of the etalon was 3.90°, the peak wavelength was 3534.9 nm, and the linewidth was 1.27 nm (1.02 cm⁻¹), which was the narrowest linewidth of a BGSe OPO, to the best of our knowledge. The beam quality was also improved after inserting the FP etalon.

Keywords: optical parametric oscillation; narrow linewidth; BaGa₄Se₇.

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1. Introduction

The tunable narrow-linewidth mid-infrared (MIR) and far infrared (FIR) lasers have important application value in many fields, such as atmospheric environment monitoring, material diagnostics, and high-resolution spectroscopy. The optical parametric oscillator (OPO) is an attractive approach, especially when high energy and average power are demanded simultaneously. The oxide-based crystals KTiOPO₄ (KTP) and KTiOAsO₄ (KTA) are well known and have often been used for generating MIR pulses in the OPO. In 2020, Lidar backscattered signals in the spectral band of 3.30–3.50 μm have been measured and analyzed along the horizontal path in the atmosphere^[1]. The tunable MIR laser was generated by KTA and KTP OPOs. The KTA crystal, as the isomorph of KTP, has many merits similar to those of KTP, such as high transparency range (0.35–5.3 μm), high optical damage threshold (450 MW/cm²), and high nonlinear coefficient ($d_{24} = 3.64$ pm/V). However, the small tuning range of the output wavelength affects its application. When using angle tuning, the output wavelength of the KTA ($\theta = 90^\circ$, $\varphi = 0^\circ$) OPO under type II (A) phase matching is less than 3.5 μm. When using temperature tuning, the slope of the output wavelength with temperature is only 0.073 nm/°C^[2].

BaGa₄Se₇ (BGSe) is a new MIR nonlinear monoclinic crystal, which was first synthesized in 2010^[3,4], to the best of our knowledge. The crystal has wide transmission range (0.47–18 μm), large effective nonlinear coefficient ($d_{11} = 24.3$ pm/V, $d_{13} = 20.3$ pm/V), high damage threshold (557 MW/cm²), and large slope of output wavelength with temperature 3.20 nm/°C^[5]. The angle tuning range and temperature tuning range of BGSe are higher than those of KTA. Yang *et al.*^[6,7] demonstrated a tunable picosecond optical parametric amplifier (OPA) with wavelength ranges of 3–5 μm and 6.4–11 μm based on the BGSe in 2013 and 2015, respectively. In 2016, Kostyukova *et al.*^[8] used a 1064 nm Nd:Y₃Al₅O₁₂ (Nd:YAG) laser to pump the BGSe OPO, and an ultra-broad tunable range of 2.7–17 μm was obtained, with a maximum pulse energy of 3.7 mJ at 7.2 μm. In 2018, Kolker *et al.*^[9] reported on the first, to the best of our knowledge, BGSe nanosecond OPO pumped by a Q-switched Nd:YLiF₄ laser at 1053 nm, and an ultra-broad tunable range of 2.6–10.4 μm was obtained. In 2018, Zhao *et al.*^[10] demonstrated a tunable 8–9 μm OPO with a type II phase-matching BGSe pumped by a 2090.7 nm acousto-optical Q-switched Ho:YAG laser, and a spectral bandwidth of 67.8 nm at 8925.7 nm was achieved. A continuous-wave MIR radiation from difference frequency generation by mixing a continuous-wave Ti:sapphire

laser and a continuous-wave YAG laser in BGSe was demonstrated in 2019^[11]. The output linewidth was 16.4 cm^{-1} (40 nm) when the idler wave was $5.1 \text{ }\mu\text{m}$. With a signal wave produced by the BGSe OPO serving as seed, the ZnGeP_2 (ZGP) OPA was demonstrated in 2020. The corresponding bandwidth (FWHM) of the signal and idler was approximately 8 and 7 nm with a peak wavelength of 3.95 and $4.44 \text{ }\mu\text{m}$, respectively^[12]. In 2020, Zhang *et al.*^[13] reported a high-energy, narrow spectral linewidth MIR laser from a Nd:YAG pumped BGSe OPO. The output linewidth of 12 nm was obtained at 3816 nm.

Compared with KTA, although BGSe has many advantages, its output linewidth is slightly larger than that of KTA^[14]. So, the BGSe OPO needs to narrow the linewidth before it can be better applied in atmospheric environment monitoring and other fields. Up to now, we have not seen the report of narrowing linewidth of BGSe OPOs. We narrowed the linewidth of the BGSe OPO by inserting a Fabry–Perot (FP) etalon into the branch of the L-shaped cavity. When the linewidth of the oscillating signal wave in the OPO was narrowed by the FP etalon, the idler wave with a narrow linewidth was obtained.

2. Methods

When the wavevector mismatch Δk is equal to zero, the pump wave, signal wave, and idler wave in BGSe achieved perfect phase matching. Under the condition of small-signal gain, the conversion efficiency of the three-wave interaction can be derived from Eq. (1):

$$\eta = \eta_0 \left[\frac{\sin\left(\frac{\Delta k}{2} L\right)}{\frac{\Delta k}{2} L} \right]^2, \quad (1)$$

where η_0 is the conversion efficiency of the perfect phase matching, L is the length of the BGSe, and Δk is the wavevector mismatch. When $\Delta k = 0$, conversion efficiency η reaches the maximum. When $\Delta k = |\pi/L|$, $\eta = 4\eta_0/\pi^2 = 0.406\eta_0$; the conversion efficiency decreases to about 40% of the maximum, but it can still be considered as effective phase matching.

When a 15 mm long BGSe (56.3° , 0°) was pumped by a 1064 nm laser under type I phase matching, the wavelength of idler wave was 3637 nm at $\Delta k = 0$ ^[15]. The wavelength of the idler wave was 3630 nm at $\Delta k = \pi/L$, and the wavelength of the idler wave was 3643 nm at $\Delta k = -\pi/L$ under the same conditions. Therefore, the acceptance linewidth of BGSe (56.3° , 0° , $L = 15 \text{ mm}$) pumped by a 1064 nm laser under type I phase matching was $3643 - 3630 = 13 \text{ nm}$. The result was consistent with the calculation result given by the equations in Ref. [16], but our algorithm is more concise.

The transmittance curve of the etalon has many peaks. When the FWHM of the peak is narrower than that of the input light spectrum, the effect of narrowing the laser linewidth can be achieved. When the inclination of the etalon changes, the peak wavelength will also be changed. The wavelength acceptance linewidth of the BGSe OPO can be converted to the frequency acceptance linewidth $\Delta\nu_{\text{BGSe}} = c \cdot \Delta\lambda/\lambda^2 = 294.83 \text{ GHz}$. In

order to make the BGSe OPO with etalon output only one peak wavelength in the idler band, the frequency interval of etalon $\Delta\nu$ should be greater than or equal to $\Delta\nu_{\text{BGSe}}$. So, the thickness of the etalon h should be less than or equal to 0.35 mm when the material of the etalon is fused silica ($n_{\text{etalon}} = 1.45$ at $1.50 \text{ }\mu\text{m}$).

The target output linewidth of the BGSe OPO after inserting the etalon in our work is 3 nm, corresponding to the $\Delta\nu'_{\text{BGSe}} = c \cdot \Delta\lambda'/\lambda^2 = 68.04 \text{ GHz}$. The transmission linewidth $\delta\nu$ should be less than or equal to $\Delta\nu'_{\text{BGSe}}$. So, the finesse of the etalon F should be greater than or equal to $\Delta\nu/\delta\nu = 4.33$. When the flatness is $\lambda/10$, the reflectivity $R \approx 0.7$ under the condition of $F = 4.33$. In light of these conclusions, a fused silica etalon with thickness $h = 0.35 \text{ mm}$, $R = 0.7$ (in the 1450–1550 nm band), and the flatness $\lambda/10$ is selected to narrow the linewidth of the BGSe OPO.

When the linewidth of signal light (in the 1450–1550 nm band) was narrowed in the OPO, the output linewidth of idler light (in the 3393–3997 nm band) would be narrowed at the same time.

3. Experiment Setup

The experimental setup is shown in Fig. 1. The BGSe OPO was pumped by an SL800 Series pulsed Nd:YAG laser with 13 ns pulse width (FWHM), 8 mm beam diameter, and 1 Hz pulse repetition frequency. A diaphragm (D) was placed behind the Nd:YAG for adjusting the light path, and the beam diameter was compressed from 8 mm to 4 mm through a telescope system (T, BX-1064-2X) to improve the energy density of the pump wave.

The BGSe crystal is a $6 \text{ mm} \times 8 \text{ mm} \times 15 \text{ mm}$ cuboid with a cutting angle of (56.3° , 0°). BGSe crystals were finely polished and AR coated at the pump, signal, and idler wavelengths.

M1 is highly transmissive (HT) for the pump (P, $T > 95\%$) and highly reflective (HR) for the signal (S, $1.4\text{--}1.6 \text{ }\mu\text{m}$, $R > 99\%$). M2 is a 45° flat mirror, HT for the pump (P, $T > 95\%$), HR for the signal (S, $1.4\text{--}1.6 \text{ }\mu\text{m}$, $R > 99\%$), and HT for the idler (I, $3.4\text{--}4.3 \text{ }\mu\text{m}$, $T > 95\%$). M3 is HR for the signal (S, $1.4\text{--}1.6 \text{ }\mu\text{m}$, $R > 99\%$).

E is a $15 \text{ mm} \times 15 \text{ mm} \times 0.35 \text{ mm}$ fused silica etalon (Corning 79800a) coated with 70% AR film at 1450–1550 nm on both sides, with the flatness of $\lambda/10$ (Fujian CASIX), and placed in the optical path at a small angle. The etalon was fixed on the outside of the round hole of the upper adjusting frame (Zolix NMV12.7), and the knob of the adjusting frame was used to adjust the tilt angle.

Fig. 1. Schematic diagram of the experimental setup.

A filter (F) and a Ge plate (G) were placed behind M2. The transmittance of the F is about 1% at 1064 nm and 95%–99% at 3–5 μm . The transmittance of Ge is zero at 1064 nm and about 60% at 3–5 μm . The idler wave was detected by a grating spectrometer (GRA, Omni- λ 300, Zolix). The idler wave transmitted from the GRA was measured by a detector (DEC, PCI-10.6 from Vigo). The idler wave energy from PCI-10.6 was measured by a DSOX3054 oscilloscope (OSC). When the maximum energy emerges in the OSC, the wavelength set by the GRA is the peak wavelength of the idler wave.

4. Result and Discussion

The output spectrum of BGSe (56.3°, 0°) pumped by 1064 nm under type I phase matching is shown in Fig. 2, while the ambient temperature was about 14°C.

Fig. 2. Output spectrum of BGSe.

As shown in Fig. 2, the black dots were the experimental data of the idler wave when the output pulse energy of the idler wave was 0.49 mJ. When the transmission wavelength of the GRA was set to 3529 nm and 3530 nm, the average voltage recorded on the OSC was 184.52 mV and 208.71 mV, which were the two maximum data. Therefore, the peak wavelength was between 3529 nm and 3530 nm. When the transmission wavelength of the GRA was set to 3527–3532 nm, the OSC had a response. When the transmission wavelength of the GRA was set outside 3524–3532 nm, the OSC did not respond. According to the experimental data points, the Gaussian curve can be used to fit. The Gaussian function obtained by fitting was $y = 202 \times \exp\left(-\frac{x-3529.4}{2.72}\right)^2$, where the output linewidth (FWHM) was $2\sqrt{\ln 2} \times 2.72 = 4.53$ nm. If the wavenumber was used to represent the output linewidth, it was 3.64 cm^{-1} .

According to Section 2, the acceptance linewidth of BGSe (56.3°, 0°, $L = 15$ mm, type I) pumped by 1064 nm was 13 nm, which was larger than the measured output linewidth of 4.53 nm. This may be due to the following reasons: (1) the acceptance linewidth was obtained under the small-signal gain condition, while the output linewidth was obtained in the OPOs, and the mode selection of the OPO cavity mirror reduces the linewidth of the idler wave; (2) BGSe had some absorption of parametric waves, and the OPO mirror was not fully aligned. The experiment loss of parametric waves was larger than the theoretical, especially when the phase mismatch Δk increases. Therefore, the parametric light, which can be excited in theory, will not be able to vibrate in practice.

The output linewidth of the BGSe OPO is shown in Fig. 3 when the FP etalon was inserted in the L-shaped cavity.

The black dashed line in Fig. 3 is the output spectrum fitted according to the theoretically calculated acceptance linewidth (13 nm) without etalon. The black circles represent the measured data without etalon when the output pulse energy of the idler wave was 0.49 mJ, and the black solid line is the output

Fig. 3. Output spectra of BGSe.

spectrum (center wavelength 3529.4 nm, linewidth 4.53 nm) obtained by Gaussian function fitting according to the black measured data points. The color curves 1–6 represent the output spectra of the etalon at different inclination angles after inserting the etalon. The color circles represent the measured data with the etalon inserted when the output pulse energy of the idler wave was about 0.5 mJ, and the curves of different colors were fitted by the measured data points at different inclination angles according to the Gaussian function. Color line 1 is the output spectrum of the etalon with the inclination angle of about 0°, and the color lines 2, 3, 4, 5, and 6 correspond to the increasing inclination of the etalon. The thickness and refractive index of the etalon will change slightly under the influence of ambient temperature. However, the FP etalon in the experiment was made from fused silica and had a thin thickness, so the influence of the environment was relatively small. The results are shown in Table 1.

As shown in Table 1, the peak transmittance of the etalon at normal incidence $\lambda_{FP}(m)$ was at about 3526.5 nm. According to Section 2, the previous and next transmittance peaks of the etalon were at about 3514.5 nm and 3538.5 nm, respectively, which were recorded as $\lambda_{FP}(m-1)$ and $\lambda_{FP}(m+1)$. The interval between two transmittance peaks was $\Delta\lambda_{FP}$ and $\Delta\lambda_{FP} = \lambda_{FP}(m+1) - \lambda_{FP}(m) \approx 12$ nm. When the inclination angle of the etalon increases, the transmittance curve moves to the long-wave direction, and the $\lambda_{FP}(m)$ was changed to $\lambda_{FP}(m, \alpha)$, where α was the inclination of light outside the etalon. When the angle of light outside the etalon was 3.19°, the peak wavelength increased by 6 nm, and $\lambda_{FP}(m, 3.19^\circ) = 3532.5$ nm. When the angle of light outside the etalon was 4.50°, the peak wavelength increased by 12 nm, and the peak wavelength $\lambda_{FP}(m, 4.50^\circ) = \lambda_{FP}(m+1) = 3538.5$ nm.

In the experiment, six curves were measured between the external inclination angles of 0° and 3.90°. The theoretical inclination angles of light outside the etalon were 1.45 times the theoretical inclination of light in the etalon ($n_{\text{etalon}} = 1.45$). The measured inclination angles were a little bigger than the theoretical inclination angles of light outside the etalon. This might be caused by the machining error of the etalon or the vibration of the FP etalon in the experiment.

The peak wavelength of the BGSe OPO represented by color line 4 was 3529.2 nm when the measured external inclination angle of the etalon was 2.34°, which was broadly consistent with the peak wavelength of the BGSe OPO output spectrum without the etalon. The linewidth was narrowed from 4.53 nm to 2.05 nm, and the peak voltage also increased. This is because the energy of the narrow-linewidth laser was more concentrated. When the inclination of the etalon changed, the peak voltage was also changed. This is caused by noise or unstable output idler light energy, and no certain law was observed in the experiment.

The output peak wavelength of the BGSe OPO without the etalon was 3529.4 nm at 14°C, which was recorded as λ_{BGSe} . λ_{BGSe} varied with the ambient temperature and incident angle of the pump wave. There were three cases on λ_{BGSe} and $\lambda_{FP}(m)$.

- (1) $\lambda_{BGSe} = \lambda_{FP}(m)$. The peak wavelength of the BGSe OPO was exactly equal to the peak wavelength of the transmittance curve of the FP when the inclination was 0°. The linewidth of the BGSe OPO can be narrowed without adjusting the etalon's inclination.
- (2) $\lambda_{BGSe} > \lambda_{FP}(m)$ and $|\lambda_{BGSe} - \lambda_{FP}(m)| < (\Delta\lambda_{FP})/2$. The inclination of the etalon can be adjusted to make $\lambda_{FP}(m, \alpha) = \lambda_{BGSe}$ achieve the same peak wavelength before and after linewidth narrowing. At this time, the angle of adjustment was small, and the theoretical angle of light outside the etalon was 0°–3.19°.
- (3) $\lambda_{BGSe} < \lambda_{FP}(m)$ and $|\lambda_{FP}(m) - \lambda_{BGSe}| < (\Delta\lambda_{FP})/2$. The inclination of the etalon can be adjusted to make $\lambda_{FP}(m-1, \alpha) = \lambda_{BGSe}$ achieve the same peak wavelength before and after linewidth narrowing. At this time, the angle of adjustment was a little larger, and the theoretical angle of light outside the etalon was 3.19°–4.50°.

Therefore, the peak wavelength of BGSe OPO output can be consistent before and after linewidth narrowing by adjusting the inclination angle of the etalon in this experiment, and the adjustment range of the inclination angle was within 4.50°. Due to the transmission of the etalon, when the inclination angle was larger, the cavity loss was larger, and the pump threshold was higher. In addition, if the peak wavelength of the BGSe OPO before and

Table 1. Output Wavelength and Linewidth of BGSe OPO with Etalon at Different Inclination Angles.

No. of Colored Lines	Peak Wavelength (nm)	Theoretical Inclination of Light in Etalon (°)	Theoretical Inclination of Light Outside Etalon (°)	Measured Inclination of Light Outside Etalon (°)	Linewidth (FWHM) (nm)
1	3526.5	0	0	0	1.80
2	3526.7	0.40	0.58	0.78	1.52
3	3527.6	0.95	1.38	1.56	1.57
4	3529.2	1.46	2.12	2.34	2.05
5	3531.7	2.04	2.96	3.12	1.58
6	3534.9	2.59	3.76	3.90	1.27

Fig. 4. Comparison of the output beam quality factor with and without the etalon.

after linewidth narrowing need not be consistent, the output wavelength of the BGSe OPO can be tuned by adjusting the angle of the etalon. The difference between the peak wavelength of the BGSe OPO before and after inserting the etalon was $0-\Delta\lambda_{FP}/2$.

After inserting the etalon, the pump threshold increases slightly due to the increase of intracavity loss. The pump threshold of the BGSe OPO was about 16.1 mJ without the etalon. The pump threshold of the BGSe OPO was about 18.8 mJ with the etalon (the measured inclination of light outside the etalon was 2.34°), which was slightly higher than that without the etalon. The slope efficiencies were 11.61% without the etalon and 11.53% with the etalon, which was basically consistent.

As shown in Fig. 4, the beam quality was improved after inserting the FP etalon. The inset in Fig. 4 shows the corresponding spatial profile near the focus of the lens ($f = 300$ mm). The beam sizes were measured by a pyroelectric array camera (Ophir-Spiricon PY-III-HR) at different distances. With the hyperbolic curve fitting method, the M^2 values of the BGSe OPO without the etalon were measured to be 3.13 and 2.42 in the horizontal and vertical directions, respectively. When the FP etalon was inserted in the L-shaped OPO, the M^2 values were measured to be 2.56 and 2.03 in the horizontal and vertical directions, respectively. We think that the improvement of longitudinal mode quality will improve the quality of the

transverse mode at the same time. However, this still needs further research.

5. Conclusion

The linewidth of the BGSe OPO was narrowed in an L-cavity by inserting an FP etalon for the first time, to the best of our knowledge. The theoretical acceptance linewidth was 13 nm when BGSe (56.3° , 0° , $L = 15$ mm) was pumped by a 1064 nm laser under type I phase matching. The experimental linewidth of the output linewidth without the etalon was 4.53 nm (peak wavelength 3529.4 nm). After inserting the fused silica FP etalon with thickness $h = 0.35$ mm, $R = 0.7$ (in the 1450–1550 nm band), and the flatness $\lambda/10$, the peak wavelength changed from 3526.5 nm to 3534.9 nm, and the linewidth decreased to 1.27–2.05 nm when the inclination angle of the etalon increased from 0° to 3.90° . When the inclination angle of the etalon was 2.34° , the linewidth was 2.05 nm, and the peak wavelength was still 3529.2 nm. The pump threshold of the BGSe OPO with the etalon was slightly higher than that without the etalon, while the beam quality was improved after inserting the etalon.

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