Diode-end-pumped solid-state ultraviolet laser based on intracavity third-harmonic generation of 1.06 μm in YCa₄O(BO₃)₃ crystal

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Abstract

Intracavity type-I sum-frequency mixing of 1.06 μm and 532 nm with a (θ, φ) = (106°, 77.2°)-cut YCOB crystal was performed in a compact laser-diode-pumped solid-state laser. Three type-II phase-matching KTP crystals with different length were used to generate 532 nm light by frequency-doubling of 1.06 μm. The 355 nm output power was measured with the three KTP crystals for Q-switched and continuous-wave (CW) operation, respectively. The maximum ultraviolet output power of 1305 μW was obtained with a 15 mm KTP crystal for CW operation, while the maximum ultraviolet average output power of 124 mW was obtained with a 10 mm KTP crystal for Q-switched operation.

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1. Introduction

In recent years, short wavelength coherent radiation in ultraviolet (UV) region has attracted wide attention for a variety of applications such as machining, spectroscopy, optical data storage, laser printing, undersea communication and medical treatment. Nonlinear optical (NLO) conversion of solid-state lasers operating in the near-infrared range is currently a very effective method for UV light generation. Compared to traditional ultraviolet gas lasers, diode-pumped solid-state ultraviolet lasers have many advantages, including compactness, high efficiency, long lifetime, high stability and all solid-state construction. Nowadays LiB₃O₄ (LBO) and β-BaB₂O₄ (β-BBO) are the most widely used nonlinear optical crystals for UV laser-beam generation. However, these crystals suffer from some sort of limitations, such as the difficulty of growth, hygroscopy, small acceptance angle and large walk-off. Therefore it is very necessary to search for new NLO crystals which can mitigate these limitations.

During the last few years, newly developed NLO crystals of ReCa₄O(BO₃)₃ (ReCOB) (Re = Y, Gd) have attracted much attention due to their good optical properties [1–6]. They possess many advantages such as non-hygroscopy, large nonlinear coefficient which is comparable to that of LBO, high damage threshold, wide transmission band, large phase-matching range and good mechanical properties allowing easy polishing. What is more, since they melt congruently, they can be grown rapidly and easily to large-size single crystals with high optical quality by using conventional Czochralski technique. While other famous NLO crystals, such as KTP, KDP, LBO and β-BBO, do not possess this advantage. So ReCOB has been recognized as a promising NLO crystal in the frequency-conversion domain.

Until now, most of the research interest involving ReCOB has been focused on the aspects of second-harmonic generation (SHG) and self-frequency doubling (SFD). In 1997, Iwai found that YCOB crystal can reach type-I phase-matching for third-harmonic generation (THG) of 1064 nm by means of sum-frequency mixing (SFM) (1064 + 532 → 355 nm), while GdCOB cannot [1]. The transmission limit of YCOB can be as short as 200 nm, while for GdCOB there are several sharp absorption peaks in the region of 200–320 nm. So YCOB is more suitable for UV light generation than GdCOB. Recently, our group have fitted the spatial distribution curve of dₑₑₑ (effective NLO coefficient) for the THG of 1064 nm in YCOB according to its second-order NLO susceptibilities. By the calculating and the extracavity THG experiments, we found that the...
largest \( d_{\text{eff}} \) for THG of 1064 nm is near the direction of \((\theta, \phi) = (106^\circ, 77.2^\circ), [7]\).

In this paper, we report a compact diode-pumped solid-state ultraviolet laser, in which a \((106^\circ, 77.2^\circ)-\)cut YCOB crystal has been used for type-I sum-frequency mixing of 1.06 \( \mu \)m and 532 nm to generate 355 nm light. The 532 nm light was generated by SHG of 1.06 \( \mu \)m with type-II phase-matching KTP crystals. Two kinds of NLO process, SHG and SFM, occurred in one resonant cavity, which was very complicated. We investigated the 355 nm output power with three KTP crystals with different length as frequency-doubler for continuous-wave (CW) and Q-switched operation, respectively.

2. Experimental setup

The conversion efficiency of THG depends on efficient SHG significantly. In our previous intracavity SHG work [5,8,9], a three-mirror folded resonator can generate efficient second-harmonic. So the intracavity THG experiment was carried out in the similar resonator configuration, as shown in Fig. 1. A fiber-coupled laser diode with center wavelength around 808 nm was employed as the pump source in the intracavity THG experiment. The laser crystal, a 0.6 at\%, a-cut \( 4 \times 4 \times 7 \) mm\(^3\) Nd:YVO\(_4\), was AR coated at 808 nm and 1.06 \( \mu \)m on the pump face, and AR coated 1.06 \( \mu \)m on the opposite face. M\(_2\), a flat mirror with a dual-wavelength HR coating at 1.06 \( \mu \)m and 532 nm on its inside surface, was mounted on a translation stage. So the mode size in the laser crystal could be changed conveniently because the length of M\(_2\)M\(_3\) arm determined it significantly. M\(_2\), whose transmission at 355 nm was measured to be 87.3\%, served as the output coupler for the THG light. M\(_3\), a concave mirror with radius of curvature of 100 mm, was HR coated at 1.06 \( \mu \)m and HT coated at 355 nm on the curved surface, AR coated at 532 nm on the other surface. So in the THG experiment, there is also green laser radiation through M\(_3\) simultaneously. A filter \((T = 76\% \text{ at } 355 \text{ nm}, T < 1\% \text{ at } 1.06 \mu \text{m}, 532 \text{ nm})\) was placed between M\(_2\) and the power meter to block the first- and the second-harmonic laser beams. The acousto-optical (A-O) Q-switch was placed close to Nd:YVO\(_4\) crystal.

The \( 5 \times 5 \times 11 \) mm\(^3\) YCOB crystal, AR coated at 1.06 \( \mu \)m and 532 nm on both end faces, was \((106^\circ, 77.2^\circ)-\)cut for type-I SFM of 1.06 \( \mu \)m and 532 nm. It was placed closed to the end mirror M\(_2\) where a beam waist located, because high power density in NLO crystal is necessary for efficient NLO conversion. And the KTP crystals, which were all AR coated at 1.06 \( \mu \)m and 532 nm on both end faces, were also placed closed to YCOB crystal for efficient SHG. The lengths of the three KTP crystals were 5, 10 and 15 mm, respectively.

3. Experimental results

The CW operation of intracavity THG was performed by removing A-O Q-switch firstly. We optimized the length of M\(_2\)M\(_3\) arm to 75 mm by translating M\(_2\). Because the output power of 355 nm with 5 mm KTP crystal was too low to measure, Fig. 2 only shows the results obtained with 10 and 15 mm KTP crystals. The corresponding THG conversion efficiency as a function of incident pump power is shown in Fig. 3. From these figures, we can see that the ultraviolet output power and THG conversion efficiency obtained with 15 mm KTP was higher than that obtained with 10 mm KTP crystal. It was due to the higher SHG conversion efficiency obtained with 15 mm KTP crystal. The conversion efficiency was almost linearly proportional to incident pump power. At the incident pump power of 4.05 W, the maximum ultraviolet-light output power was measured to be 1305 and 614 mW with 15 and 10 mm KTP crystals, respectively. And the corresponding pump-to-ultraviolet conversion efficiency were 0.032\% and 0.015\%. At the same time, we could also obtained 375 and 62 mW of 532 nm radiation through M\(_3\), respectively.

Then, we performed Q-switched operation of intracavity THG with the three KTP crystals. In our preliminary experiment, the maximum ultraviolet average output power could...
4. Discussion

Based on the Sellmeier equations of YCOB crystal given in Ref. [10], the angular acceptance of the YCOB crystal used in our experiments, $\Delta \theta l$ and $\Delta \phi l$ were calculated to be 34.5 and 12.5 mrad mm for Type-I THG ($1064 + 532 \rightarrow 355$ nm), respectively. And the walk-off angle was also calculated to be 8.9 mrad, which is smaller than that of LBO [11].

Since two NLO processes, SHG and SFM occurred in one arm of the cavity, the operation in the cavity was somewhat complicated. Too many factors such as the balance of power density between the first and second harmonic in YCOB crystal [12], the polarization-matching condition and so on, determined the efficiency of the THG. In general, the power of second harmonic is quadratically proportional to the power of fundamental wave. So it is difficult to keep the balance of power density between the first and second harmonic over the whole pump range. Fig. 5 shows the polarization directions of the fundamental, second and third harmonic in the KTP and YCOB crystals. A fraction of the fundamental wave generated from Nd:YVO$_4$ crystal was converted to the second harmonic in Type-II ($\epsilon_1 + \epsilon_2 \rightarrow \epsilon_2$) KTP crystal as a “doubler”. And the unconverted fundamental wave was subsequently mixed with the second harmonic to generate the third harmonic in Type-I ($\epsilon_1 + \epsilon_1 \rightarrow \epsilon_2$) YCOB crystal as a “tripler”. Thus, the fundamental and second harmonic could be well overlapped spatially, which was advantageous for the conversion efficiency of THG.

The fundamental and second harmonic beams were loosely focused ($l < 2\pi w_0^2 n_{1,2}/\lambda_{1,2}$) in YCOB crystal corresponding to a confocal parameter ($b = 2\pi w_0^2 n_{1,2}/\lambda_{1,2}$), where $w_0$ is the beam waist radius, $n$ is the refractive index, $l$ is the length of the nonlinear crystal, $\lambda$ is the wavelength, and the subscripts 1,2 refer to the fundamental and second harmonic) of about 57 and 28 mm, respectively. For a loose-focus condition the power $P_3$ of the third harmonic is given by [13]

$$P_3 = \frac{\omega_3^2 d_{3\text{eff}}^2 P_1^2 P_2}{\varepsilon_0 c^3 n_1 n_2 n_3 (\pi w_0^2)}$$  \hspace{1cm} (1)$$

where $\omega_3$ is the frequency of 355 nm light, $\varepsilon_0$ is the permittivity of free space, $c$ is the vacuum speed of light, $d_{3\text{eff}}$ is the effective NLO coefficient, the subscripts 1, 2 and 3 refer to the 1064, 532 and 355 nm beams, respectively. So the power of the third harmonic is proportional to that of the fundamental wave and second harmonic. From Fig. 5, we can see that a fraction of fundamental wave did not take part in the SFM process due to the type-II SHG and type-I SFM. It is also one of the reasons for the low THG efficiency.

Another reason could be that the 355 nm light was generated by the single pass of 1.06 $\mu$m and 532 nm beams in YCOB crystals because most of the 532 nm light (over 300 mW at the maximum pump power) was radiated through M3. Moreover, since the beam propagation directions of the three YCOB crystals were cut slightly away...
from the phase-matching direction, YCOB crystals, not AR coated at 355 nm, were somewhat tilted, which could cause additional cavity losses. Therefore, we think that the THG efficiency can be increased significantly by some improvement of the experimental conditions.

5. Conclusions

In summary, we investigated the intracavity THG of 1.06 μm in a (106°, 77.2°)-cut YCOB crystal for type-I SFM, with three different-length KTP crystals as frequency-doubler, for CW and Q-switched operation, respectively. The maximum ultraviolet output power of 1305 μW was obtained with the 15 mm KTP crystal for CW operation, while the maximum ultraviolet output power of 124 mW was obtained with the 10 mm KTP crystal for Q-switched operation. Therefore, YCOB crystal is suitable for not only SFD and SHG, but also THG to generate ultraviolet light.

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