Generation of femtosecond laser pulses at 396 nm in $K_3B_6O_{10}Cl$ crystal

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$K_3B_6O_{10}Cl$ (KBOC), a new nonlinear optical crystal, shows potential advantages for the generation of deep ultraviolet (UV) light compared with other borate crystals. In this paper we study for the first time the second harmonic generation (SHG) of a femtosecond Ti:sapphire amplifier with this crystal. Laser power is obtained to be as high as 220 mW at the central wavelength of 396 nm with a 1-mm-long crystal, and the maximum SHG conversion efficiency reaches 39.3%. The typical pulse duration is 83 fs. The results show that second harmonic (SH) conversion efficiency has the room to be further improved and that the new nonlinear crystal is very suited to generate the high efficiency deep ultraviolet laser radiation below 266 nm.

Keywords: nonlinear optics, nonlinear optical crystal, second harmonic generation

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1. Introduction

Ultraviolet lasers have applications in various areas, such as time resolved spectroscopy, electronic industry, photolithography, biology, etc.[1,2] To generate UV light with high conversion efficiency, second harmonic generation (SHG) and sum frequency generation (SFG) based on the nonlinear optical (NLO) technology are often applied to visible and infrared (IR) solid-state lasers. In particular, NLO crystal plays a key role in the efficient laser frequency conversion process. As such, the search for new crystals with excellent second order NLO response attracts persistent attention. Using a $KB_5O_8{\cdot}4H_2O$ crystal, UV radiations between 217.3 nm and 234.5 nm were generated for the first time in 1975.[3] After that, a series of excellent borate crystals have been discovered, such as LiB_3O_5 (LBO),[4] β-BaB_2O_4 (BBO),[5] $CsLiB_6O_{10}$ (CLBO),[6] $KBe_2BO_3F_2$ (KBBF),[7] and SrB_4O_7 (SBO).[8] Among them, KBBF is the only crystal that can be used to generate UV light below 200 nm.[9,10] However, wide application is limited by its layered structure. In 2015, using BBSAG crystals, high repetition, tunable deep-ultraviolet pulse below 200 nm was obtained by Meng *et al.*[11] Chen *et al.* demonstrated a UV 330-nm laser in CBO crystal through SHG of Nd:YLF red laser in 2016.[12] In the same year, 266-nm UV light generated with BABF crystal was presented by Yang *et al.*[13] Recently, Wu *et al.* reported on $K_3B_6O_{10}Cl$ (KBOC) as a novel high quality NLO crystal with a size of up to 35 mm × 35 mm × 11 mm.[14] Its novel physical and optical properties exhibit a great potential for nonlinear frequency conversion in the UV spectral range.

The KBOC crystal is grown by the top-seeded solution method and possesses a perovskite-related structure. It is a negative uniaxial crystal and represents the trigonal crystal system.[15,16] Wang *et al.* studied the influence of pressure on the SHG tensor of KBOC in 2013.[17] Han *et al.*[18] and Gong *et al.*[19] investigated the electronic, elastic, piezoelectric, acoustic, and the Raman spectroscopic properties based on first-principles in 2013 and 2014, respectively. The calculated thermal conductivities of KBOC at 60 °C are 2.21 $W{\cdot}m^{-1}{\cdot}K^{-1}$ and 1.98 $W{\cdot}m^{-1}{\cdot}K^{-1}$ along *a* and *c* axes, respectively, which is larger than that of BBO along the *c* axis (1.2 $W{\cdot}m^{-1}{\cdot}K^{-1}$).[14] KBOC also has a high damage threshold of more than 9 GW/cm^2 measured by a 1-Hz, 5-ns laser system at 1064 nm. Its transmittance spectrum ranges from 180 nm to 3400 nm which is 10-nm shorter than that of BBO in the UV cut-off.

In this work, we experimentally investigate for the first time the frequency doubling of a femtosecond Ti:sapphire amplifier in KBOC. A second harmonic (SH) conversion efficiency is achieved to be as high as 39.3% with a 1-mm-long KBOC crystal and its maximum output power reaches

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220 mW at 396 nm.

2. Crystal nonlinear optical properties and experimental setup

Figure 1 shows the refractive indices of the KBOC crystal for extraordinary and ordinary light, respectively. The Sellmeier equations of KBOC are derived by Wu *et al.*[14] The birefringence is about 0.05.

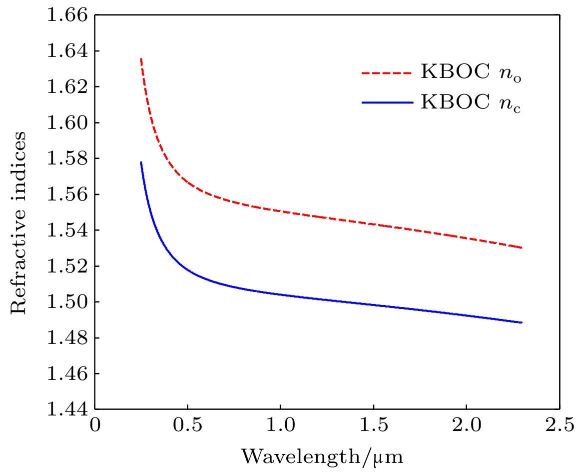

Fig. 1. (color online) Refractive indices of the KBOC crystal for extraordinary and ordinary light as a function of wavelength.

In order to understand the second harmonic properties of the KBOC crystal, we first calculate the type-I and type-II phase-matching and the corresponding walk-off angles between the fundamental frequency (FF) light (ordinary wave) and the SH light (extraordinary wave), and the results are shown in Fig. 2. On the basis of the curves, the shortest possible SH wavelengths are ~ 272 nm in type-I mode and ~ 355 nm in type-II mode, respectively. The walk-off angle varies drastically between 550 nm and 700 nm and then declines gently. The maximum walk-off angle for SHG is about 1.9° at 780 nm. The effective nonlinear coefficient is another vital NLO parameter for SHG in KBOC, which is given by Eq. (1). The respective coefficients are $d_{15} \approx 0$ pm/V and $d_{22} \approx 0.88$ pm/V at 1064 nm. The NLO characteristics of KBOC, BBO, and CLBO for type-I phase matching at 800 nm are given in Table 1. Note that the KBOC is non-hygroscopic which suggests that it could ensure stable operation for a long time. In addition, the small walk-off angle may compensate for the small effective nonlinear coefficient to yield a considerable SH conversion efficiency.

$$\begin{aligned} d_{\text{eff(ooe)}} &= d_{15}\sin(\theta) - d_{22}\cos(\theta)\sin(3\varphi), \\ d_{\text{eff(eoe)}} &= d_{22}\cos^2(\theta)\cos(3\varphi). \end{aligned} \quad (1)$$

Table 1. NLO characteristics of KBOC, BBO, and CLBO for type-I SHG at 800-nm wavelength.

Crystal	UV cut-off/nm	Phase-matching angle/(°)	Walk-off angle/(°)	d_{eff}/(pm/V)	Damage threshold/(GW/cm^2)	Hygroscopicity
KBOC	180	42.7	1.85	0.65	9[a)]	none
BBO	190	29.2[20]	3.90	2.00	13.5[b)]	slight
CLBO	180	36.2[20]	2.05	0.481	26[b)]	high

[a)] Measured by 5-ns, 1-Hz, 1064-nm laser. [b)] Measured by 1-ns, 10-Hz, 1064-nm laser.

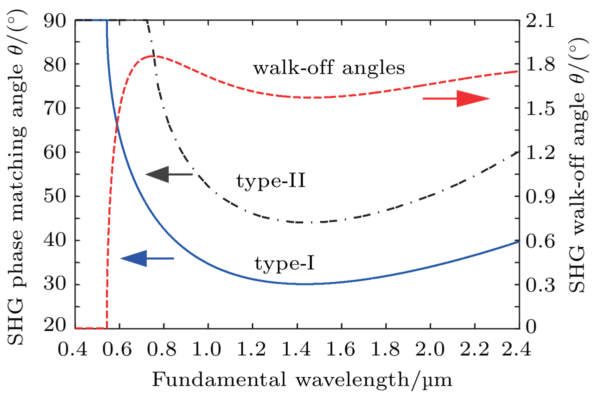

Fig. 2. (color online) Type-I and type-II phase-matching and walk-off angles for SHG in KBOC each as a function of fundamental wavelength.

The schematic diagram of the SHG experimental setup is shown in Fig. 3. It includes a commercial femtosecond Ti:sapphire chirped pulse amplification (CPA) laser at 1 kHz repetition rate, a 5:1 Galilean telescope and a KBOC crystal. The laser system provides up to 0.8-mJ pulse energy at 800 nm with a pulse duration of 27 fs. Through the Galilean telescope, the spot diameter of the incident beam in the KBOC crystal is reduced to ~ 5 mm. The SH light is separated from the FF light by a dichroic mirror (DM) which is high reflection-coated between 370 nm and 430 nm and anti-reflection-coated between 750 nm and 850 nm. In this experiment, two KBOC crystals with lengths of 2 mm and 1 mm are anti-reflection-coated for the FF and the SH. The measured transmittances are higher than 99.8% at the FF and 98.8% at the SH respectively. Both crystals have an identical aperture size of 8 mm × 8 mm and are cut at $\theta = 42.7°$ and $\theta = 30°$ for type-I phase-matching at 800 nm.

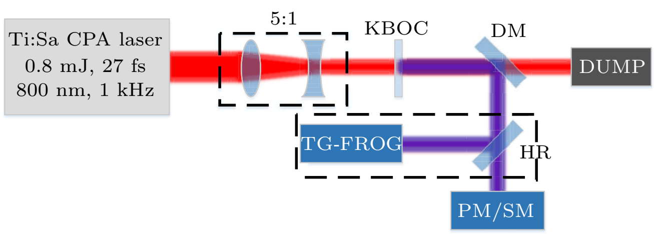

Fig. 3. (color online) Schematic diagram of the experimental setup for SHG. DM: dichroic mirror, PM: power meter, SM: spectrometer, HR: high reflection-coated mirror, TG-FROG: dispersion-free transient-grating frequency-resolved optical gating.

3. Results and discussion

The SH output power and the corresponding conversion efficiency each as a function of the FF input power are shown in Fig. 4. For the 2-mm-long KBOC crystal (dash lines in Fig. 4), the highest conversion efficiency about 36.3% was achieved at an FF power of ∼ 300 mW and for higher FF power, the conversion efficiency declines. The maximum output power is about 192 mW when the FF power increases to about 720 mW. For comparison, the output power and the corresponding conversion efficiency are also measured with the 1-mm-long KBOC crystal (solid lines in Fig. 4). Below 350 mW of FF power, the output power of the SH is slightly lower than that of the 2-mm-long KBOC crystal. For higher FF input power, the output power with the 1-mm-long KBOC crystal is higher and increases up to 220 mW at an FF input power of 720 mW. The maximum conversion efficiency for this sample is ∼ 39.3%. For both samples, the conversion efficiencies vary similarly with FF input power, which could be due to the thickness of the KBOC crystal. In order to verify this inference, we theoretically calculate the temporal walk-off length $L = \tau / \left(\left| v_{\rm FF}^{-1} - v_{\rm SH}^{-1} \right| \right)$ between FF and SH. Here, τ is the pulse duration of FF and $v_{\rm FF}$ and $v_{\rm SH}$ represent the group velocities of FF and SH, respectively. Our results indicate that the optimum thickness of the KBOC crystal in this experiment is ∼ 0.2 mm for an input pulse duration of 27 fs. With the increase in SH output power, back conversion and other NLO effects become inevitable in the thick KBOC crystal, resulting in the decline of the conversion efficiency. In turn, reducing the thickness of the crystal can effectively improve the SH output power and the conversion efficiency. Using a thinner crystal, the SH output power and conversion efficiency could be further increased.

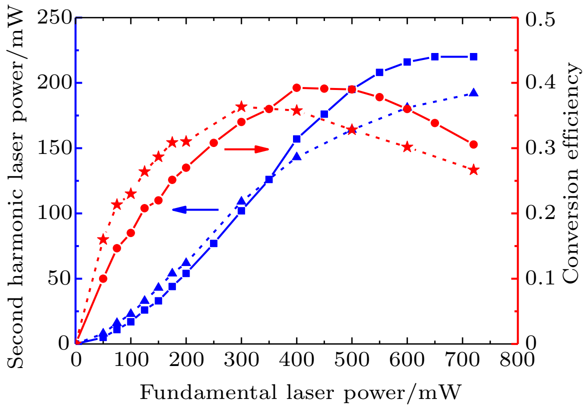

Fig. 4. (color online) Plots of SH output power and conversion efficiency versus FF input power. Solid curves with square and circular symbols represent the SH output power and conversion efficiency, obtained with a 1-mm-long KBOC, respectively. Dash curves with triangle and pentagram symbols represent the SH output power and conversion efficiency, obtained with a 2-mm-long KBOC, respectively.

We also characterize the SH pulses by a home-made dispersion-free transient-grating frequency-resolved optical gating (TG-FROG) with a 0.05-mm-thick silica as the nonlinear medium. The measured and reconstructed TG-FROG traces are shown in Fig. 5, along with the retrieved pulses in frequency and time domain, the spectrum directly measured by a spectrometer and the transform limited (TL) pulse width. The full width at half maximum (FWHM) of the measured spectrum is about 2.7 nm centered at 396 nm. The pulse duration of the SH is calculated to be 83 fs without chirp compensation while the TL pulse width corresponding the spectrum retrieved by FROG is 41.2 fs.

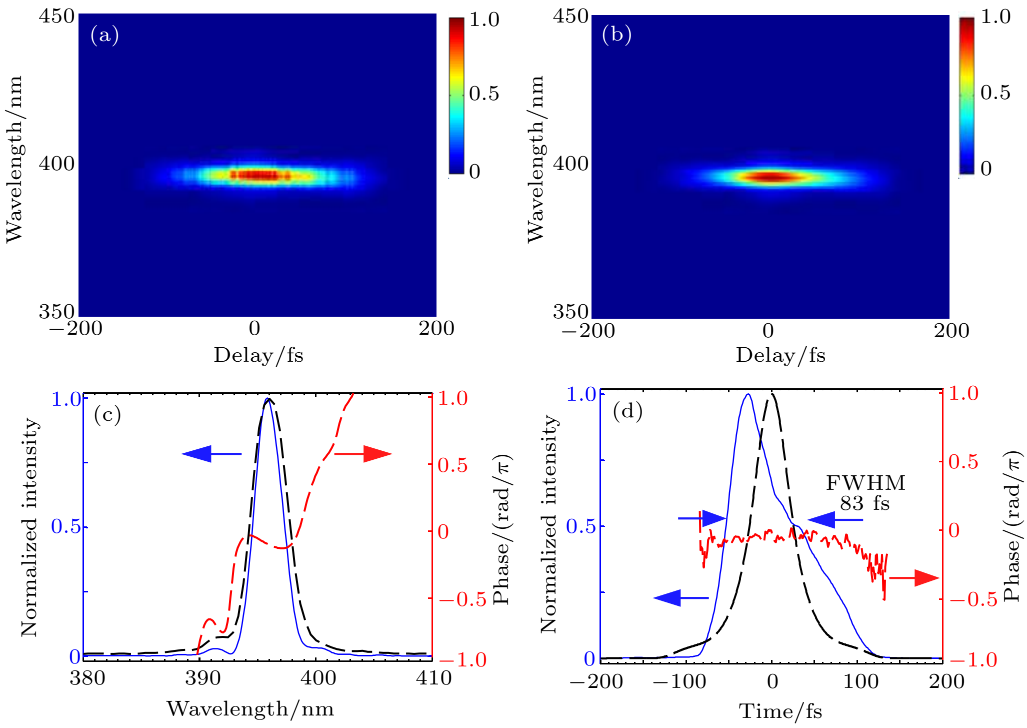

Fig. 5. (a) Measured and (b) reconstructed TG-FROG trace. Solid and dash curves represent the retrieved and directly measured spectra (c), and the retrieved and TL pulse widths (d), respectively.

4. Summary

In this work, we demonstrate the SHG from a femtosecond Ti:sapphire amplifier by using a novel nonlinear KBOC crystal. UV pulse of 83 fs at 396 nm with a maximum output power of ∼ 220 mW and a highest conversion efficiency of 39.3% are obtained in a 1-mm-long KBOC crystal. Theoretical analyses and experimental results both support that higher output power, conversion efficiency, and shorter pulse duration are all possible with thinner crystal. Considering the transmittance edge down to 180 nm, the KBOC crystal will be generally feasible to generate high power vacuum UV pulses through SHG and subsequent SFG.

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