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Impact of BaB_2O_4 growth method on frequency conversion to the deep ultra-violet

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ABSTRACT

In this article, we report how the growth method used for barium beta-borate β-BaB_2O_4 (BBO) impacts its high power second harmonic generation properties in the deep-UV. We compared a BBO crystal grown by flux (Top Seeded Solution Growth or TSSG) and a BBO crystal grown by the Czochralski (CZ) method. We first characterized their transparency properties, then we measured their single-pass second harmonic conversion efficiencies with both a low average power and a high average power nanosecond pulsed lasers. We show that both crystals have comparable linear absorption and conversion efficiencies at low power, whereas in a high power experiment, the CZ-grown BBO yields higher conversion efficiency than the TSSG grown BBO. With a 30 W, 150 kHz, 8 ns green laser, the use of a CZ BBO led at best to a 40% increase in available average output power at 257 nm.

1. Introduction

The material machining industry has continuously looked to process more and more types of materials efficiently. For some of them such as silicon, glass, sapphire, the ultraviolet lasers (UV) were developed in order to increase the material absorption [1,2]. While UV lasers around 350 nm obtained by third harmonic generation (THG) are now commonly used in material processing, deep-UV lasers obtained by fourth harmonic generation (FHG) around 260 nm are still challenging to realize. The nonlinear material quality is a key parameter in order to reach high and stable conversion efficiency by reducing thermal dephasing and degradation effects. Beta-barium borate β-BaB_2O_4 (BBO) is a nonlinear crystal that has good properties for frequency conversion to the UV, such as a high nonlinearity or a short cut-off wavelength in the UV around 190 nm [3]. It is available commercially, and is usually grown by top-seeded solution growth (TSSG) assisted with either a Na_2O or NaF flux [4]. BBO can be also grown without flux by the Czochralski (CZ) method, but it is more difficult and produces smaller crystal boules [5]. Several previous works argued that the CZ-grown BBO had better properties when used for frequency conversion to the UV. Umezu and al reported in 1998 that CZ-BBO lead to an improved laser lifetime, but without showing any material characterization [6]. In 2012, Bandhari and al reported a 3 MW peak power 266 nm microchip laser based on fluxless-grown BBO. They showed a comparison between flux-grown and CZ BBO, and demonstrated that a CZ-grown BBO had a significantly lower linear absorption than flux-grown BBO, which led to an improvement in the final conversion efficiency [7]. Nonetheless, the flux-grown BBO had an unusually high absorption value at 266 nm of $\alpha = 0.143\ cm^{-1}$. Moreover, the improvement of the BBO transparency was explained by a combination of several factors not only linked to the CZ growth method.

In this paper, we investigate if the CZ growth of BBO intrinsically leads to an improved crystal quality, and if this improved quality can lead to enhance performances during high-power fourth harmonic generation to the deep-UV at 257 nm.

2. Crystals considerations

Two crystals have been investigated during this experiment. A

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flux-grown BBO was purchased (TSSG). The Czochralski BBO (CZ) was grown at the LMOPS laboratory (Metz). We synthesized BaB_2O_4 by the following solid phase chemical reaction from standard purity raw materials, $BaCO_3$ (Alfa Aesar 99.95%) and B_2O_3 (Alfa Aesar 99.98%):

$$BaCO_3 + B_2O_3 \rightarrow BaB_2O_4 + CO_2\uparrow$$

A standard Czochralski type crystal growing apparatus with radio frequency (RF) heated furnace and without automatic diameter control was employed. The melt had to be super-cooled near 1050 °C for the nucleation of the β-phase. In our apparatus the radial temperature gradient was measured to be about 28°/cm. The vertical temperature gradient was adjusted to 550°/cm at the surface of the melt. The seed was oriented along the c axis and its bottom surface was polished and etched with ortho-phosphoric acid at 100 °C for 1 min to eliminate stress on the surface [5]. A 50 mm high platinum crucible with a diameter of 50 mm was used. The typical rotation speed is 4 rpm. With a pulling rate of 0.5 mm/h, a 58 g bowl was obtained as shown Fig. 1.

The two BBO crystals samples had a section 3 × 3 mm and a length of 3.155 mm, and were cut for critical type-I second harmonic generation from 532 nm to 266 nm at a cut angle of $\theta = 47.4°$. Their optical facets have been polished with the same setup to a roughness below 8 Å, and were not AR coated. The crystals have been kept under dry air until the experiment.

3. Absorption and second harmonic generation measurements

Their linear absorptions were measured from 190 nm to 700 nm using a Perkin Elmer Lambda 900 spectrometer. Their transmittances are displayed in Fig. 2. The UV cut-off is located at 189 nm for both crystals, and no major differences between the different transmission curves are observed. From the transmittance values, the absorption coefficients were calculated. Their values near the cut-off wavelength are displayed in the inset of Fig. 2.

From these calculations, we extracted the absorption coefficient of both crystals at 257 nm, which is equal to 0.033 cm^{-1} and 0.050 cm^{-1} for crystals TSSG and CZ respectively. We can notice several things from these measurements. First, both these values are below 0.06 cm^{-1}, which is low compared to the 0.143 cm^{-1} previously reported for flux-grown BBO [7]. In high power experiments even a small absorption can induce a thermal dephasing strong enough to lower the conversion efficiency significantly [8]. A second remark is that the CZ BBO has a slightly higher linear absorption than the flux-grown crystal, which indicates that CZ growth itself is not enough to guarantee a lower absorption than a flux grown BBO. At these levels of absorption, parameters such as the base materials purity or growing environments strongly influence the final crystal transparency significantly. These parameters can vary a lot between a commercial grown crystal and a lab-grown one.

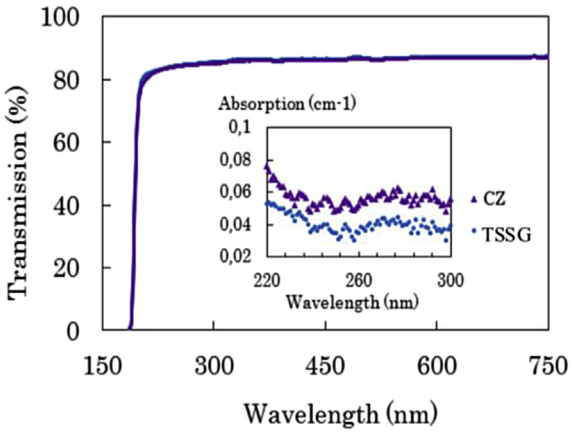

Fig. 2. Transmission curves for CZ and TSSG crystals; inset: absorption curves near 257 nm for TSSG (blue circles) and CZ (purple triangles). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

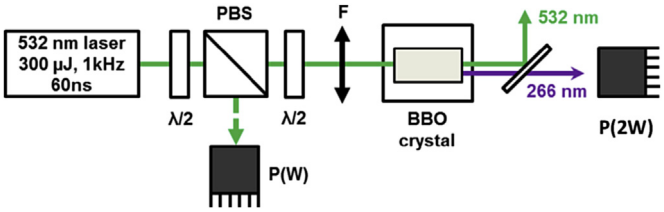

Fig. 3. Experimental setup for SHG.

In order to confirm that both BBO crystals have similar nonlinear properties, a second harmonic generation experiment at low average power was conducted. The experimental set-up is displayed in Fig. 3.

A low average power pulsed laser at 532 nm was used as the fundamental source. The laser has an average power of 300 mW at 532 nm, at 1 kHz, and a pulse width of 60 ns. It corresponds to a pulse energy of 300 µJ and peak power of 5 kW. The laser output was focused into the nonlinear crystal at a beam waist of $w_0 = 60$ µm. The crystals were controlled in temperature at 20 °C and placed on a rotation mount. The output beams were separated using a prism and an interferential filter is placed in front of detector. All the displayed values correspond to the corrected powers at the entrance and at the output of the crystal. For a maximum input power of 300 mW at 532 nm, output powers of 16.6 mW and

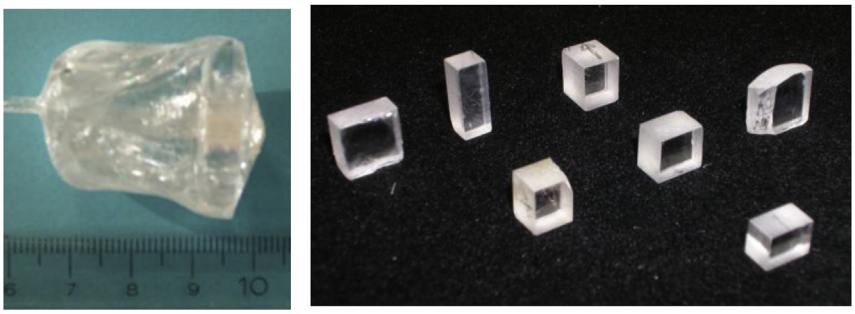

Fig. 1. Czochralski β-BBO grown in super cooled melt (left). Samples are sawn from clear and less stress part of the bowl. Oriented and polished sample (right).