Improved Growth Techniques for Nonlinear Optical Crystal CsB$_3$O$_5$ Based on the Investigation of Defects

Jingcheng Feng $^{1,2,1}$, Yuwei Chen $^{1,2}$, Feidi Fan $^{1,2}$, Heng Tu $^{1,2,*}$, Guochun Zhang $^{1,2,*}$ and Yicheng Wu $^3$

1 Beijing Center for Crystal Research and Development, Key Laboratory of Functional Crystals and Laser Technology, Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing 100190, China
2 Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, China
3 Institute of Functional Crystals, Tianjin University of Technology, Tianjin 300384, China
* Correspondence: tuheng@mail.ipc.ac.cn (H.T.); gczhang@mail.ipc.ac.cn (G.Z.)

Abstract: Growth defects in CsB$_3$O$_5$ (CBO) crystals grown using the seed-submerged growth technique (SSGT), which includes parallel grouping, growth step, and inclusion, were observed and analyzed. Dislocation was investigated using the chemical etching method for the first time. Relationships between defects and growth conditions are discussed, and ways to overcome growth defects are suggested. Using the cool-end compensation of thermocouples, a CBO crystal measuring $63 \times 40 \times 30$ mm$^3$ (weighing 190 g) was successfully grown using the SSGT. Adopting the top seeded solution growth (TSSG) method, a scattering centers-free CBO crystal measuring $75 \times 52 \times 46$ mm$^3$ (weighing 480 g) was obtained from the improved Cs$_2$O–B$_2$O$_3$–MoO$_3$ system with a molar ratio of (1–3):(1.5–3.5):(1–4), which is the largest CBO single crystal to date.

Keywords: defects; top seed solution growth; cesium triborate; nonlinear optical crystal

1. Introduction

CsB$_3$O$_5$ (CBO) crystals exhibit excellent ultraviolet nonlinear optical (NLO) performance, which has a relatively large NLO coefficient ($d_{14} = 1.49$ pm/V), a wide ultraviolet (UV) transparency range ($167$–$3400$ nm), and a high laser damage threshold ($26$ GW/cm$^2$ for $1.0$ ns pulses at $1053$ nm) [1,2]. Using CBO crystals, the maximum output power of a $355$ nm laser was up to $103$ W through the third harmonic generation (THG) of an Nd:YAG laser at $1064$ nm [3]; a stable output of $31.5$ W at $355$ nm was also achieved with a standard deviation of $1.4\%$ within five hours [4]. Both theoretical predictions and experiments have confirmed that the output power and conversion efficiency of $1064$ nm laser radiation to its THG ($355$ nm) in CBO crystals are higher than those in commercially available LiB$_3$O$_5$ (LBO) crystals under the same experimental conditions [5], which makes CBO a competitive candidate for THG. In addition, a $278$ nm laser of $10.3$ W was successfully realized by the fourth harmonic generation (FHG) of an Nd:YAG laser at $1112$ nm [6], which locates in the solar-blind UV region ($200$–$300$ nm) with high photon energy. This is the most attractive laser source for atmospheric remote sensing, communications, and molecule detection. These results indicated that CBO is a promising NLO crystal for frequency conversion in the UV region.

As CBO crystals melt congruently at $835^\circ$C and crystallization of the CBO phase was confirmed to take place in the composition range from $67$ mol % to $81$ mol % B$_2$O$_3$ [7], many growth techniques could be used to grow CBO single crystals. These growth techniques include the Czochralski method [8], the Kyropoulos method (stoichiometric melt [8] and $8.9$ mol % Cs$_2$O-enriched melt [9]), the seed-submerged growth technique (SSGT) (3–5 mol % Cs$_2$O-enriched melt [10]), and the top seeded solution growth (TSSG) method (higher than $16.7$ mol % Cs$_2$O-enriched melt [11,12], NaF flux [13], and Cs$_2$O–MoO$_3$ flux [14,15]). CBO crystals grown via the Cs$_2$O–MoO$_3$ flux system using the TSSG method...
were considered to have high optical quality, but the growth period was too long, and it took over 60 days to grow an approximately 220 g crystal [15]. It took approximately one week to grow a 160 g crystal using the SSGT [10], but repeatability was lower overall, and the crystal quality was poor, which was mainly related to growth defects. To date, only a few reports regarding CBO crystals’ defects are available [16–19]. Therefore, it is important to investigate CBO crystals’ growth defects to facilitate growing crystals with large size and high quality in a short period.

In this study, CBO crystals grown using the SSGT were used to investigate growth defects, including parallel grouping, growth step, inclusion, and dislocation. Some effective ways to overcome these growth defects were suggested. Eventually, a single CBO crystal measuring $75 \times 52 \times 46$ mm$^3$ (480 g in weight) was successfully grown via the improved $\text{Cs}_2\text{O–B}_2\text{O}_3–\text{MoO}_3$ system using the TSSG method.

2. Materials and Methods

2.1. Crystal Growth

CBO crystals for investigating growth defects were grown using the SSGT. The molar ratio of $\text{Cs}_2\text{O}:\text{B}_2\text{O}_3$ was 1.04:3, that is, an excess of 4 mol % $\text{Cs}_2\text{O}$ was added to the stoichiometric melt. A vertical resistance heated furnace with three zones, each of which was independently controlled by a Shimaden FP23 controller with an accuracy of $\pm 0.1 ^\circ\text{C}$, was adopted to achieve the desired negative temperature gradient in the axis direction. Consequently, the temperature of the solution’s surface was approximately $1 ^\circ\text{C}$ higher than the temperature 10 mm beneath of the solution’s surface, where a (101) direction seed crystal measuring $2 \times 2 \times 1$ mm$^3$ attached to the end of $\phi 1.5$ mm platinum wire was slowly introduced. The cooling rate, rotating rate, and growth period were 0.1–0.3 $^\circ\text{C}/\text{day}$, 15–30 rpm, and 1–2 weeks, respectively. The specific growth process was the same as that described in [10]. The grown crystals were used in the following defect studies.

2.2. Chemical Etching Experiment

Dislocations in CBO crystals were investigated using the chemical etching method. The transparent and macro-defect-free parts of CBO crystals were fabricated into wafers measuring $6 \times 6 \times 5$ mm$^3$ along the (100), (010), and (001) directions, respectively, of which the two cross-sections $6 \times 6$ mm$^2$ were precisely polished. At room temperature, the polished wafers were immersed in a glycerol solution with concentration of 50% for 5 min. Etched wafers were observed under the Nikon 104 metallographic microscope with a magnification of 400:1.

3. Results and Discussion

3.1. Parallel Grouping

During the growth of CBO crystals, parallel grouping frequently appeared, as shown in Figure 1a. There were two main reasons for parallel grouping. The first was spontaneous nucleation, which resulted from foreign impurities, including volatiles and furnace materials. When spontaneous nucleation grew to a certain size, it touched the growing crystal and eventually formed a parallel grouping crystal. In addition, with the continuous volatilization of the solution at high temperature, the growth system became rich in $\text{B}_2\text{O}_3$ and viscosity increased, which tends to trigger spontaneous crystallization. The second reason for parallel grouping was a change in undercooling due to a sudden change in temperature of the growing system or external environment. This resulted in abrupt changes and local imbalances in growth rates, which led to the emergence of parallel grouping crystals. To avoid parallel groupings, several effective methods have been implemented, such as controlling the cooling rate and maintaining a stable external growth environment to avoid mutation via undercooling; adopting a suitable raw material ratio to reduce the volatilization and viscosity of the system; and purifying the environment and using cleaner furnace materials to decrease foreign impurities.
3.2. Growth Steps

During the initial growth stage, the growing crystal was transparent. However, growth steps appeared when the crystal grew to a certain size (as shown in Figure 1b); this was primarily related to the temperature gradient. If there is an unstable and uniform temperature gradient in the growth region, when the growth boundary expands outward, there are differences in supersaturation at various locations around the growing crystal. The greater the temperature difference, the easier it is for growth steps to occur. In addition, the mutation of undercooling can also lead to growth steps. Maintaining a stable temperature gradient and precisely controlling the cooling rate can effectively reduce growth steps.

3.3. Inclusion

As shown in Figure 1c, there were some inclusions in the CBO crystal. Melt diffusion was not sufficient during the growth process due to the high viscosity of the growth system, and different supersaturations in the melt contacted the growing crystal. Generally, there is more supersaturation at the corners and edges of a growing crystal than in the center, which leads to faster growth at the edges and corners than the center, resulting in a concave center and convex circum. In an extreme case, the crystal grows to be dendritic-like. If the growth rate is later reduced, the surface can become flat again, trapping the melt inside and forming inclusions. Inclusions can be reduced or eliminated by adjusting the initial components and their ratios to decrease the viscosity of the melt and maintain a stable temperature field. Moreover, in a CBO crystal’s early growth stage, appropriately increasing the rotation rate can accelerate the substance transport in the melt and make the diffusion more uniform, which helps reduce inclusions.

3.4. Dislocation

Chemical etching has proved to be one of the most powerful methods for revealing the nature and distribution of dislocations. To obtain appropriate etching conditions, glycerol solutions with concentrations of 30–90% were tested, and etching time was varied from 5 min to 3 h. When etching time was extended, the crystal’s surface dissolved and etching pits could not be clearly observed. Results indicated that 50% was a suitable concentration of glycerol aqueous solution, with an etch time of approximately 5 min at room temperature. Figure 2a–c show microscopic images of different direction CBO wafers etched using 50% glycerol aqueous solution for 5 min at room temperature. Generally, the symmetry of an etch pit is in accordance with the symmetry of the crystal face, and the shapes of etch pits on different faces are different for the same crystal [20]. In this study, etch pits on the (100) plane were helical-like, which reflected the 21 axis along the (100) direction in the CBO crystal, whereas etch pits on the (001) plane exhibited a rectangular pyramid, indicating the orthorhombic symmetry of the CBO crystal. The etch pit densities (EPDs) on the (100) and (001) planes were calculated as $7.48 \times 10^3$ and $1.57 \times 10^3$ cm$^{-2}$, respectively [21], which were less than the reported values of the $\beta$-BaB$_2$O$_4$ crystal [22]. However, no etch pits were observed on the (010) plane, because water molecules tend to permeate the CBO crystal along the a axis direction [23]. Selecting high quality seed crystal, avoiding spontaneous
nucleation, reducing foreign impurities, and minimizing thermal stress are beneficial in reducing dislocation in as-grown crystals.

Figure 2. The etch pits on (a) (100) plane, (b) (001) plane, and (c) (010) plane of a CBO crystal.

3.5. Improved Growth Techniques for CBO Crystal

According to the investigation and analysis of the above defects, it was found that the SSGT could be used to grow CBO crystals by adjusting temperature distributions, selecting appropriate cooling and rotation rates, reducing foreign impurities, and so on. Most importantly, to obtain a stable external growth environment, the cold-end compensation of the thermocouple was especially necessary. The so-called cold-end compensation is carried out by placing the cold end of the thermocouple in an ice-water mixture to ensure the consistency of the cold-end temperature and reduce fluctuation in the growth temperature caused by changes in environmental temperature. In the improved growth experiment, the cold-ends of the growth furnace’s three thermocouples were placed in an ice-water mixture throughout the growth period. High-purity Cs2CO3 (915 g) and H3BO3 (1002 g) were mixed and melted in a φ 95 × 80 mm3 platinum crucible in several batches. The temperature was decreased by 0.5 °C in a 14-day growth period. Other conditions and detailed procedures are described in Section 2.1. The as-grown 190 g CBO crystal measuring 63 × 40 × 30 mm3 is shown in Figure 3a; its crystallographic planes were determined using X-ray diffraction, as sketched in Figure 3b. This was the heaviest CBO crystal grown using the SSGT; its lower part was completely transparent, and no inclusions could be observed with the naked eye.

Figure 3. (a) CBO crystal grown using the seed-submerged growth method and (b) sketched morphology.

As the platinum wire was embedded in the grown crystal, the upper part of the crystal inevitably cracked, which greatly reduced the crystal’s utilization rate. In addition, owing to the high viscosity and volatility of the growth system, there were still some scattering centers in the as-grown CBO crystal, which was similar to CBO crystals grown via the self-flux system using the TSSG method. These scattering centers were related to the crystalline precipitate nucleated during the cooling process after crystal growth. Post growth heat treatment with quenching and the vapor transport equilibration (VTE) process...
can effectively reduce or eliminate scattering centers; however, post-treated crystals were prone to crack, and had lower laser damage thresholds than as-grown crystals [7,17–19]. Fortunately, the TSSG using the Cs₂O–MoO₃ flux system has been proven to successfully grow scattering-free CBO crystals [24]. However, the previous Cs₂O–MoO₃ flux systems have a lower growth temperature (~650 °C) and a longer growth period (60–90 days). Therefore, we improved its component ratio by increasing the concentration of solute CBO and adjusting the Cs₂O/MoO₃ ratio of the flux. The increase in solute concentration shortened the growth period and increased the growth temperature, which also helped reduce the growth system’s viscosity. The appropriate molar ratio of Cs₂O:B₂O₃:MoO₃ was confirmed to be in the range of (1–3):(1.5–3.5):(1–4). In a practical growth experiment, high-purity raw materials (Cs₂CO₃, H₃BO₃, and MoO₃) with a 3282 g combined weight were homogeneously mixed and melted in several batches in a 120 mm diameter, 100 mm high platinum crucible. Detailed procedures and conditions can be found in [15]; however, the saturation temperature was raised to approximately 730 °C and the cooling rate was increased to 0.7 °C day, which compressed the growth period to approximately 50 days. Eventually, a 480 g CBO crystal measuring 75 × 52 × 46 mm³ was obtained, as shown in Figure 4a, which did not show any scattering centers under the irradiation of a 532 nm laser with 5 mW. To our knowledge, this was the largest CBO single crystal to date. Its schematic morphology was determined based on diffraction indices, as depicted in Figure 4b, and was consistent with the result predicted based on the PBCs theory [25]. Obviously, CBO crystals grown using (100) seed crystal exhibit well-developed large (101) and (011) faces, which are coincident with that in [26]. This long columnar crystal significantly benefited the processing of crystal devices and increased the crystal’s utilization rate.

![Figure 4](image-url)  
**Figure 4.** (a) CBO crystal grown using the TSSG method and (b) schematic morphology.

Two CBO crystal blocks with 6 × 6 × 10 mm³ dimensions along the c axis were cut from the as-grown crystals and then polished for spectroscopic measurements. The transmission spectra in the spectral range of 185–3000 nm were measured using a Lambda 900 spectrophotometer, as shown in Figure 5. The results indicated that the CBO crystals grown using two methods both had a transmittance greater than 80% and no obvious absorptions in the 200–2000 nm range. Two crystal wafers with 8 × 8 × 3 mm³ dimensions along the optical axis were processed for optical homogeneity measurements at 633 nm using a WYKO RTI 4100 interferometer. The optical homogeneities characterized by the root-mean-square of the gradient of the refractive index reached 8.816 × 10⁻⁶ and 5.946 × 10⁻⁶, for the crystals grown using the seed-submerged growth method and TSSG method, respectively (Figure 6). The CBO crystal grown using the TSSG method exhibited
a relatively higher optical quality, and its laser-induced damage threshold (LIDT) was determined using a longitudinal single mode Q-switched Nd:YAG laser at 1064 nm. The laser operated at a 1 Hz repetition rate with a 5 ns pulse duration. The 150 μm diameter beam was focused into the samples and the LIDTs were recorded using the 1-on-1 test procedure. The pulse fluence corresponding to a damage probability value of 20% was defined as the LIDT of the specimen; the LIDT was determined to be 5.66 GW/cm², which was higher than previously reported values (4.50 and 4.97 GW/cm²) under the same conditions [24]. These results suggested that the as-grown CBO crystals had high optical qualities and that the improved growth techniques were reasonable.

![Transmission spectra of CBO crystals grown via the seed-submerged growth method and the TSSG method.](image)

**Figure 5.** Transmission spectra of CBO crystals grown via the seed-submerged growth method and the TSSG method.

![Optical homogeneity values of CBO crystals grown via the seed-submerged growth method and the TSSG method.](image)

**Figure 6.** Optical homogeneity values of CBO crystals grown via the (a) seed-submerged growth method and (b) the TSSG method.

### 4. Conclusions

The growth defects of the CBO crystals grown using the SSGT, including parallel grouping, growth step, inclusion, and dislocation, were investigated. Parallel groupings were primarily caused by spontaneous crystallization and mutation as a result of undercooling. Growth steps were associated with the stability of the temperature field. Inclusions were triggered by high viscosity and inadequate diffusion of the melt. Dislocations in the CBO crystals mainly occurred on the (100) and (001) planes, and etch pit density (EPD) was calculated to be 7.48 × 10³ and 1.57 × 10⁵ cm⁻², respectively. The above defects were effectively eliminated or reduced by adjusting the temperature gradient, maintaining a stable and clean external growth environment, controlling cooling and rotation rates, optimizing the components and their proportions in the growth system, using cool-end compensation for thermocouples, and so on. For the SSGT, using 4 mol % Cs₂O-enriched melt, it was vital to adopt the cold-end compensation for thermocouples to grow large CBO crystals. For the TSSG method, the Cs₂O–B₂O₃–MoO₃ system with a molar ratio of (1–3):(1.5–3.5):(1–4) was favorable for growing CBO crystals without scattering centers; this was beneficial
for reasonably increasing the growth temperature, reducing the system’s viscosity, and shortening the growth period. In addition, optical homogeneity values \((8.816 \times 10^{-6} \text{ and } 5.946 \times 10^{-6})\), transmission spectra, and LIDT \((5.66 \text{ GW/cm}^2)\) measurements indicated the as-grown crystal’s high optical quality. To date, the TSSG method using Cs\(\text{S}_2\O\text{MoO}_5\) as flux has been proven to be the optimal growth method for large scattering centers-free CBO crystals; this will be beneficial in promoting the application of CBO crystals in the field of UV frequency.

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