Large Ti-doped sapphire bulk crystal for high power laser applications

G. Alombert-Goget a, K. Lebbou a,⇑,⁎, N. Barthalay b, H. Legal b, G. Chériaux c

a Institut Lumière Matière, UMR5306 Université Lyon 1-CNRS, Université de Lyon, 69622 Villeurbanne cedex, France
b RSA Le Rubis SA BP 16, 38560 Jarrie/Grenoble, France
c LOA, ENSTA – Ecole Polytechnique, UMR 7639, Chemin de la Hunière, 91761 Palaiseau Cedex, France

A R T I C L E   I N F O

Article history:
Available online 4 February 2014

Keywords:
Ti-doped sapphire
Large single crystals
Kyropoulos technique
Laser

A B S T R A C T

Large Ti-doped sapphire single crystals have been successfully grown by Kyropoulos technique for optical amplification. Without post growth annealing, the absorption and emission spectra do not show any presence of unwanted impurities. The obtained results indicate the possibilities to get crystals with homogeneous Ti3+ ion concentrations in large sections using this growth technique.

© 2014 Elsevier B.V. All rights reserved.

1. Introduction

The excellent mechanical, thermal, and optical properties of Ti-doped sapphire (Ti-Al2O3) allow the development of laser systems with unprecedentedly high average and peak powers [1]. Ti-sapphire has the widest relative emission bandwidth (Δλ/λ) which allows a very broad laser wavelength tunability extending from about 660 to 1180 nm, and the production of the shortest laser pulses, below 10 fs [2,3]. As a matter of fact, researchers have already demonstrated petawatt peak power laser operation using large diameter Ti:Al2O3 crystals [4]. Such result and future ones at even larger scales necessarily require large size, ultra-high-quality crystals (free of any defects) with relatively high dopant concentrations. Therefore, emphasis was placed on the development and improvement of several crystal growth technologies to produce high quality Ti-doped sapphire crystals. Attempts have been made to scale up the Czochralski [5,6], HEM [4] and Kyropoulos [7] processes to grow large boules but the inherent properties of Ti-doped sapphire make it difficult to produce them with a high enough defect-free optical quality. Among the problems encountered with Ti-doped sapphire there is its anisotropy and the smaller distribution coefficient of Ti3+ ion in the grown crystal [8], which is very detrimental to the production of large laser quality single crystals. As the titanium (effective) segregation coefficient differs from unity (K = Cs/Cl < 1), both a high proportion of (Ti3+) active ions and a good homogeneity in their spatial distribution are key factors to obtain good quality Ti:Al2O3 laser crystals. Today, the challenge for the laser crystal engineers and the titanium–sapphire users is to build the largest possible crystal boules while keeping optimal mechanical, thermal and optical properties.

In this study, we have succeed to grow large Ti-doped sapphire crystals by Kyropoulos technique. The optical characterization and laser measurement realized showing some promising results to increase the crystal diameter simultaneously control the distribution of longitudinal and transversal titanium concentrations and keep high optical qualities.

2. Experimental part

Ti3+-doped sapphire single crystals were grown by the Kyropoulos technique (KT). The details of the growth process have been published in our previously paper [7]. At room temperature, the absorption spectra in the range 200–800 were recorded. The most important point for growing homogeneous Ti-doped sapphire crystals is the control of titanium concentration in the melt and the crystal. Glow discharge mass spectroscopy (GDMS) analysis was used to measure the titanium concentration. In addition, Ti3+ qualitative measurement of the grown crystals was inspected by microluminescence analysis. The crystal was placed under a microscope (lens ×100) and was directly excited with the focused beam of a 532 nm frequency-doubled and Q-switched Nd:YAG laser. The luminescence was recorded through the microscope by an optical fiber and transmitted to a monochromator that dispersed the emitted light onto a CCD detector cooled to 0 °C.

3. Results and discussions

3.1. Crystal characterization

Fig. 1 shows Ti-doped sapphire crystal. The crystal was transparent and colored because of titanium doping. The diameter of the crystals was 100 mm, and the weight was about 6 kg, depending on the size of the crucibles and of the starting raw materials.
The crystal was grown from the melt with titanium ion concentration in the range 0.15–0.25 at%. It is transparent and colored because of titanium doping level, exempt of cracks, inclusions, and other scattering centers. Quite often, defects such bubbles were observed in undoped and Ti-doped sapphire [9–11], but in this work, the crystals were grown under stationary stable regime, they do not contain macroscopic and microscopic bubbles defects which is a great economically advantage to exploit a large area of the grown crystals. Fig. 2 shows the absorption spectra of the Ti-doped sapphire crystal, a broad absorption band was observed in the visible region with a peak at 490 nm and a shoulder at about 520 nm. The luminescence lifetime of Ti$^{3+}$ is 3 μs at room temperature.

Microluminescence spectra were recorded on the wafers with diameter of 15 and 40 mm and thickness of 20 mm respectively cut from the grown ingot. The microluminescence measurements allow a qualitative characterization of the Ti$^{3+}$ spatial distribution inside the crystals. The radial distribution of Ti$^{3+}$ in the Ti-doped Al$_2$O$_3$ crystal expressed as luminescence intensity at 730 nm is illustrated in Fig. 3. The 15 mm and 40 mm diameter samples exhibit a high homogeneous radial Ti$^{3+}$ distribution. In previous works, usually the radial distribution of titanium concentration presents a relatively large gradient at the periphery of the crystal [7], while the significant result in this study demonstrates that we get a high degree uniform titanium distribution in large size Ti–sapphire crystals.

3.2. Optical qualities

The optical qualities of the samples were evaluated by absorption measurement and wavefront transmission measurements (Fig. 4). The optical characteristics of the 15 mm and 40 mm diameter crystals are presented in Table 1.

The absorption measurements manifest that both the small and large diameter crystals possess excellent absorbance property and absorption coefficient for 526 nm laser. Both two crystals obtained desirable high FOM values, and the FOM value of 15 mm diameter crystal (200) is a little bit higher than the 40 mm diameter crystal (150). The wavefront transmission measurements indicate that the wavefront profile of 15 mm diameter crystal is perfectly plane (rms 15 nm) while which of 40 mm diameter crystal contains some correction of the figure (rms 172 nm), that is to say the former crystal is almost defects free while the latter contains some microscope defects which may depress the optical quality of the crystal, this result is consistent with the FOM values of the two crystals.

3.3. Optical amplification

The crystals have been optically pumped by a frequency-doubled Nd:YAG laser emitting 800 mJ of 532 nm light at a repetition rate of 10 Hz for the 15 mm. The 40 mm diameter crystal has been pumped by Nd:glass, 10 J and 0.1 Hz. The titanium doped sapphire crystals were positioned in a multi pass amplifier of standard chirped pulse amplification (CPA) laser chain. For the small diameter crystal, the seed pulses to be amplified have energy of 8 mJ. An
amplified energy of 300 mJ after 4 passes has been obtained. The result is consistent with numerical calculations of amplification. The second crystal has allowed obtaining 1.5 J after two passes in the amplifier with a 22 mm laser beam diameter. The laser beam shape is shown in Fig. 5. This confirms that the doping of the sapphire host was completely adequate and in good agreement with the crystal quality.

4. Conclusions

Large and high optical quality Ti$^{3+}$-doped sapphire crystals have been successfully grown by the Kyropoulos technique. Without any post growth annealing, the microluminescence spectra indicate the homogeneity of Ti$^{3+}$ concentrations in the grown crystal. Two different diameters of 15 mm and 40 mm wafers were studied for laser amplification. Both two crystals have a high intrinsic as grown FOM values (>100). The absorption and transmission measurements manifest the 15 mm diameter crystal is exempt of any defects. These high quality crystals permit to obtain good optical amplification results. The obtained results are promising for the growth of large Ti-doped sapphire crystals by Kyroupolos technology for PW scale amplifiers.

References