Impact of growth atmosphere on langatate (La$_3$Ga$_{5.5}$Ta$_{0.5}$O$_{14}$) crystals grown by czochralski technique and its coloration

B. Boutahraoui [a], E.A. Ghezal [b], A. Nehari [c], K. Zaidat [d], M. Allani [e], H. Cabane [f], M. Dumortier [f], I. Gerasymov [g], O. Sidletskiy [g], S. Obbade [h], L. Jouffret [i], K. Lebbou [c,*]

[a] Université Blida 1, BP 270 Route de Soumâa, Blida, Algeria
[b] Laboratory of Organic Chemistry and Natural Substance, ZIANE Achour University, Djelfa, Algeria
[c] Institut Lumière Matière, UMR5306 Université Lyon 1-CNRS, Université de Lyon, Lyon, 69622, Villeurbanne Cedex, France
[d] Univ. Grenoble Alpes, Grenoble INP, SIMAP-UMR CNRS, F-38000, Grenoble, France
[e] FEMTO-ST Institute, UFC, CNRS, ENSMM, UTBM, 25000, Besançon, France
[f] Cristal Innov – Université Lyon 1, 73800, Sainte-Hélène du Lac, France
[g] Institute for Scintillation Materials, National Academy of Sciences of Ukraine, 61072, Kharkiv, Ukraine
[h] Univ. Grenoble Alpes, Univ. Savoie Mont Blanc, CNRS, Grenoble INP, LEPMI, 38000, Grenoble, France
[i] Université Clermont Auvergne, CNRS, Sigma Clermont, Institut de Chimie de Clermont-Ferrand, Campus des Cézeaux, 24 Avenue Blaise Pascal, TSA60026-S60026, 63178, AUBIERE CEDEX, France

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ABSTRACT

Langatate (LGT) crystals of La$_3$Ga$_{5.5}$Ta$_{0.5}$O$_{14}$ composition of diameter 50 mm were grown from the melt by Czochralski technique. Using (1–2 wt %) Ga$_2$O$_3$ excess in the starting charge and growing crystal in mixture argon (0.1–1%O$_2$) gas atmosphere are a good condition to crystallize LGT under stationary stable regime. The LGT crystals grown along Z-axis exhibit strong faceting. The grown crystals were exempt of inclusions, cracks and secondary phases. The presence of oxygen in the growth chamber is necessary to limit gallium oxide evaporation and strongly affect the crystals coloration and the transmission spectra in the range (200–500 nm). The electrical resistivity is sensitive to the oxygen content in the growth environment.

1. Introduction

Langatate (La$_3$Ga$_{5.5}$Ta$_{0.5}$O$_{14}$) called LGT, retain piezoelectricity near it's melting point around 1470 °C, has higher piezoelectric coupling than quartz, and have proven acoustic-weaves (AW) temperature-compensated orientations around room temperature [1–4]. The LGT material is no pyroelectric and do not present phase transition below its melting temperature [1,5–7]. Additionally, LGT crystal has been shown to have temperature-compensated orientations for BAW, SAW, vibrating beams [3,8–12] and low-acoustic losses, achieving higher Q$_f$ (quality factor) than quartz [1,13]. The LGT crystals have a trigonal structure belonging to the point group 32-space group *P*321. There are four kinds of cation sites presented by the A$_3$BC$_3$D$_2$O$_{14}$ formula where A and B are located on a decahedral site coordinated by eight oxygen and an octa-hedral site coordinated by six oxygen, respectively. While C and D were located in tetrahedral sites coordinated by four oxygen, in the case of LGT crystals, La^{3+}occupies the A sites, Ga^{3+} fully occupies the C and D

sites, and Ta^{5+}occupy the B sites, respectively [14].

LGT crystal can be directly grown from the melt by conventional Czochralski method [10,15–19]. Several companies and laboratories have grown large LGT crystals size, however, it did not yet meet the wide commercial production requirements. The main reason for limiting this commercialization was the inhomogeneous properties of the crystal because of the evolution of the composition during crystal growth and the poor production efficiency. This is connected to the evaporation of Ga$_2$O$_3$ oxide, which generate cracks propagation [10,15,16]. Some amount of oxygen in the growth chamber is necessary to prevent gallium oxide evaporation, which leads to the non-stoichiometric crystal [20].

The coloration of LGT crystal differs from colorless to yellow-orange or pale-green depending on the growth atmosphere and thermal annealing conditions. As a function of the growth atmosphere, the crystallization of LGT crystals can be realized from La$_3$Ga$_5$Ta$_{0.5}$O$_{14}$) stoichiometric composition [21,22] or from rich gallium oxide (Ga$_2$O$_3$) concentration [10,16] to compensate the loss induced by the

* Corresponding author.
E-mail address: kheirreddine.lebbou@univ-lyon1.fr (K. Lebbou).

evaporation of gallium oxide.

Crystals grown under different atmospheres have significantly different colors: those grown under Ar [23,24] and N$_2$ [25] are colorless and exhibit optimal optical properties, while those grown under Ar + O$_2$ are orange [26–28] and all shades of yellow [24,29]. In Refs. [24,30], authors have suggested that oxygen in the atmosphere was responsible of crystals coloration. According to D.A.Spassky et al. [31], the color centers in LGT are due to not only oxygen vacancies, but also cation vacancies and charge transfer between cations. It has been found that the growth atmosphere influences the structure, coloring and optical properties of LGT crystals. Therefore, characterization and understanding of crystal defects are a fundamental requirement for the growth of high quality crystals suitable for reproducible acoustic wave's applications.

In this paper, as a function of the oxygen content we present the impact of growth atmosphere on the LGT stability and coloration. We have first grown different LGT crystals under stationary stable regime and discussed the defects propagation and the crystals coloration.

2. Experimental

Langatate material was elaborated from a mixture of high purity (4 N and 5 N) La$_2$O$_3$, Ga$_2$O$_3$, and Ta$_2$O$_3$ powders, which was mixed and pressed into flat pellets, then calcined at 1200 °C for 5 h. After this thermal treatment the composition of the mixture shown that the langatate being the dominant phase. The full synthesis of LGT takes place during the pre-melting in alumina crucible. The Langatate single crystals were grown by the conventional RF-heating Czochralski technique [10] in iridium crucible with internal diameter 90 mm and 90 mm height. The used atmosphere is a mixture of argon (0.1–1%O$_2$). The seed is oriented along Z ⟨0001⟩ axis. The iridium after-heater was used to decrease the axial temperature gradient [15].

The LGT crystals grew in an automatic diameter control Czochralski furnace. The starting materials were loaded into the iridium crucible and melted completely at around 1510 °C. The basic principle of the growth technique appears to be simple, in a first approach, the procedure is quite intuitive. The required amounts of chemicals are mixed and molten in Ir crucible, placed in thermal insulation (ZrO$_2$) in the furnace. The load kept molten for a while until chemical and thermal stability are reached. During the growth process, the melting and crucible temperature were followed by using optical pyrometer. Before the seeding, the temperature was reduced progressively and manually near the melting temperature (1510 °C) of the crystal, ideally just above its melting point 1515 °C. The seed connection to the liquid was important to confirm good melting temperature. The mass capture during connection step was around 1 g. Liquid temperature higher than LGT melting temperature will be accompanied by mass decreasing until seed disconnection from the melt. The Ir seed holder was employed to keep the seed on one side and the other side with the alumina rod. Once the thermal stabilization was reached, the seed was pulled up with the suitable pulling rate 1–1.5 mm/h and rotation rate 9–15 rpm, respectively. After the growth was completed, the crystal was cooled down to room temperature at a rate of 20–25 °C/h. The optical transmission characterization of ≈3 mm thick plane-parallel Z-cut plates optically polished on both sides was measured in the range 200–800 nm on a PerkinElmer Lambda 900 spectrophotometer. The collected spectra were analyzed using Lambda 900 software. The electrical resistivity was measured through the current intensity (l) crossing the sample at voltage (U) of 50 V using a pico-ampere meter. The LGT crystals shape was a disk of 15 mm diameter and 0.5 mm thickness (e) with a gold electrodes deposited (S) on the two polished faces.

3. Results and discussion

3.1. Starting raw materials analysis

The raw materials were prepared from a mixture of La$_2$O$_3$, Ta$_2$O$_5$, and Ga$_2$O$_3$ powder with 99.99% purity in stoichiometric proportion. In order to compensate the evaporation of Ga$_2$O$_3$ during the crystal growth process, 1–2 wt% extra Ga$_2$O$_3$ was added to the starting charge. Although the sintering temperature (1200 °C) was close to the melting point, the reaction of raw materials did not induce a complete reaction and the formation of pure LGT phase. We found three phases, LaGaO$_3$, LaTaO$_4$ and Ga$_2$O$_3$ that coexist with the LGT corresponding to non-equilibrium state.

3.2. Crystal growth

Whatever the growth atmosphere (Ar, Ar + O$_2$ (0.5%)), growing crystal from stoichiometric LGT composition without Ga$_2$O$_3$ excess was unsuccessful. The grown crystals were opaque, polycrystal (Fig. 1), contain cracks and inclusions corresponding to the strong volatility of Ga$_2$O$_3$ oxide (Fig. 2). The gallium oxide decomposition was accompanied by the LGT composition change in the La$_2$O$_3$–Ga$_2$O$_3$–Ta$_2$O$_5$ ternary equilibrium diagram [32] causing LaTaO$_3$ secondary phase precipitation and cracks propagation. The LGT Czochralski growth requires careful selection of the melt composition, so starting from a stoichiometric melt without Ga$_2$O$_3$ excess results in poor crystal homogeneity and defects. An another hand, growing LGT crystals using argon atmosphere with oxygen mixture ≥1% was accompanied by iridium deposit on the crystal surface (Fig. 3a), crystal cracking and inclusions propagation along the growth direction (Fig. 3b, c). In addition, some voids were visible inside the grown crystal. Through a serial of growth experimental, we have optimized the growth atmosphere to control the growth process (Table 1). Pulling crystals along Z<0001> direction from the melt using 1-2 wt% excess of Ga$_2$O$_3$ oxide under Ar or Ar + O$_2$ (0.5%) allowed growing transparent crystals under stationary stable regime without defects such cracks and inclusions (Fig. 4). The crystals grown in argon atmosphere were transparent and colorless, but under mixed gas (Ar + O$_2$) were orange and the coloration contrast depend on O$_2$ concentration. The first step of growth is manual. Once 2 mm of crystal was pulled, the automatic program was launched in order to control the diameter. The cone growth took around 12 h to reach the cylindrical diameter that was approximately 120-degree cone angle. The cone growth was stable until the crystal diameter reached the cylindrical diameter (50 mm). At the end of the cone growth, we did not observed instability and diameter increasing. It could be due to the stable flat interface. During all the process, we have the ability to control the growth process through the weighing of the crystal and the crucible. Fig. 5 shows the evolution of the LGT crystal and crucible weight during the growth process, which is comfortable to control the LGT crystallization and reach the stable regime. During the crystallization step, no fluctuation anomalies of the diameter was observed and a linear evolution of the mass with a steady state growth. The chemical percentage use for 50 mm diameter crystal was about 70% of the starting melt with the approximate total weight of 1100 g. The crystals were free of cracks and inclusions. The LGT crystals grown along the [0 0 0 1] direction exhibit very strong faceting, generally a nearly hexagonal shape with six well defined {1 0 0} facet planes on the side, and a (0 0 1) facet at the bottom of the boule (Fig. 6). Quite often, in the large langatate crystals grown from the melt, temperature fluctuation near growth interface causes interface instability leading to periodic striations propagation along growth direction. Such defects were observed in LGT crystals grown by C. Klemenz et al. [33]. They have a periodicity (spacing) of approximately 5 μm. In our crystals, we did not observed striations defects. The optimizations of the gas atmosphere and Ga$_2$O$_3$ composition for such growth are very important for the repeatability and yield of the crystal.
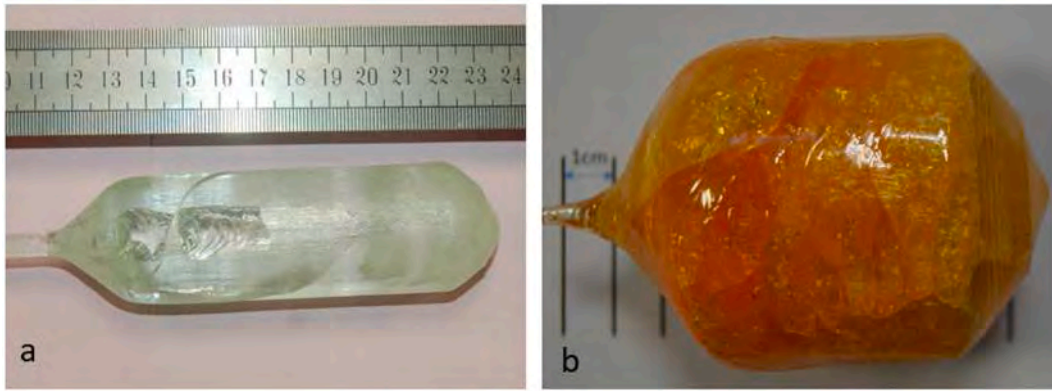
Fig. 1. Stoichiometric LGT crystal starting charge (a) LGT crystal grown in argon atmosphere and (b) grown in argon mixed 0.5%O2. For both cases we didn't' used Ga_2O_3 excess.
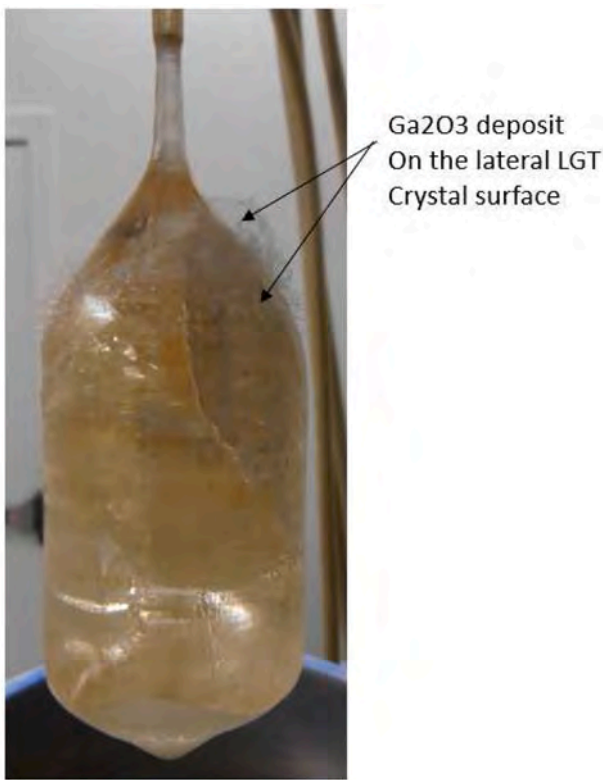

Fig. 2. Ga_2O_3 oxide deposited on the LGT surface during crystal growth from stoichiometric melt.

3.3. Optical characterization

Fig. 7 shows the transmission spectra from 200 to 800 nm of LGT crystals. All the grown crystals slightly have around 80% of transmission in the wavelength range (700–800 nm) confirming that the crystals have good optical quality. The optical transmission spectra of the crystals grown in mixed gas atmosphere significantly differ from the transmission spectra of the crystals grown in pure argon. A spread of the intrinsic absorption edge was observed as well as strong absorption at wavelengths λ in the ranges (270–300), (330–370), and (470–0.49) nm. A pronounced absorption edge around 275 nm was observed for crystal grown in mixed gas atmosphere (0.1%, 0.3% and 0.5%O_2). In case of the colorless crystal grown in argon gas, it was observed only low intensity wide band at the region 270–300 nm. The absorption band around 330–340 nm is more pronounced for crystal grown in pure Ar atmosphere and could be attributed to oxygen vacancies ($V_{\ddot{O}}$) in good agreement with references [34,35]. The intensity of this band decrease with oxygen concentration increasing and nearly disappears for crystal grown in argon (1%O_2) atmosphere. The causes of the oxygen vacancies presence in the LGT crystals grown by the Czochralski technique could be connected to the chemical composition of the starting charge and the variation in the composition of the melt during the growth process. The dissociation of the melt and evaporation of gallium suboxide result in oxygen and gallium deficiency and color contrast variation depending on the growth atmosphere.

Table 1
LGT crystals coloration as a function of the growth atmosphere.

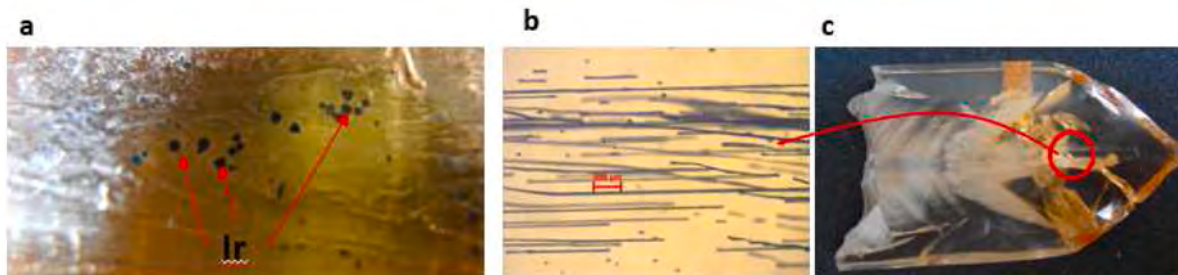| Ingot	Growth atmosphere	Color
I	Ar	Colorless
II	Ar+0.1%O_2	Orange
III-1	Ar+0.3%O_2	Light orange
IV	Ar+0.5%O2	Orange
V	Ar+1%O_2	Bright orange

Fig. 3. (a) Iridium particles in LGT crystals, (b) Inclusions in LGT crystals along the growth direction and (c) Position of inclusion analysis.
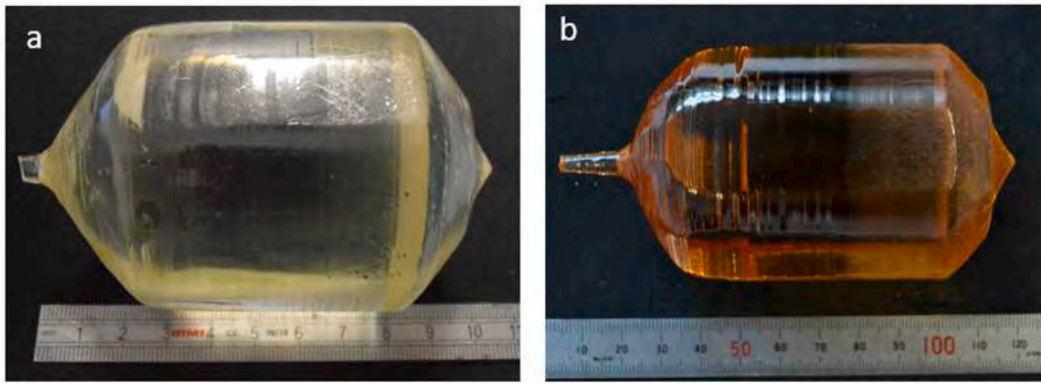
Fig. 4. LGT bulk single crystals grown by using 1% Ga_2O_3 excess: (A) Argon growth atmosphere, (B) Argon +0.5%O_2.
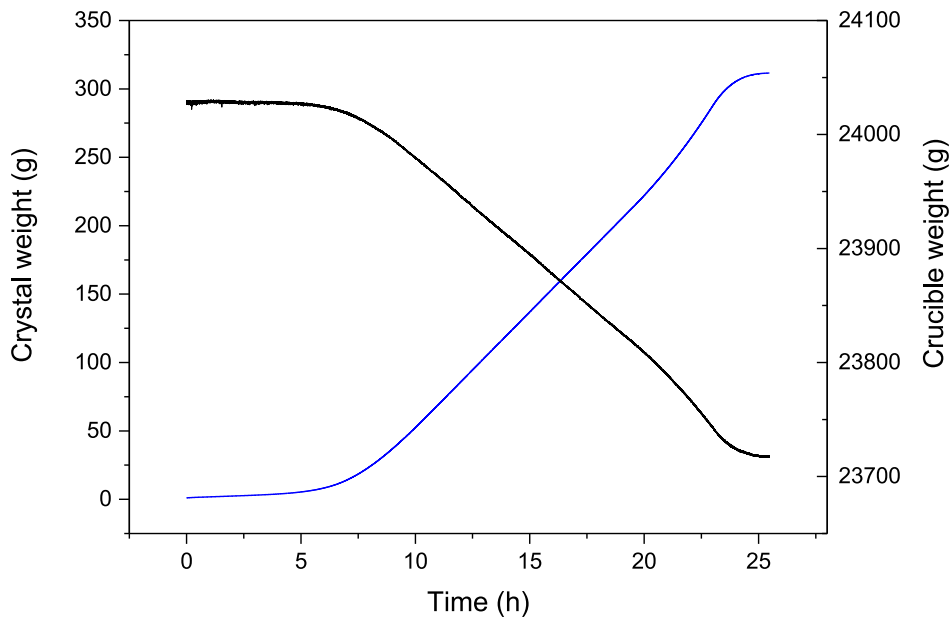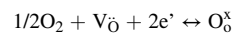

Fig. 5. Evolution of the LGT mass and the crucible weight during LGT automatic growth using 1% Ga_2O_3 excess.

Fig. 6. Facets of LGT crystal ($\varphi \approx 50$ mm) grown under stationary stable regime along Z-axis.

The oxygen content in the growing atmosphere would decrease oxygen vacancies and lead to colored crystals according to the chemical reaction equation:

$$1/2 O_2 + V_{\ddot{O}} + 2e' \leftrightarrow O_o^x$$

The absorption band around 490 can be attributed to F centers ($V_{\ddot{O}}$, 2e′) and to orange coloration in good agreement with reference [36].

Decreasing the oxygen content of the growing atmosphere significantly improves the optical properties of langatate crystals. The registered transmission spectra indicate that the addition of oxygen to Ar gas atmosphere causes a marked difference in the color of the crystals and their transmission spectral dependence. The fact that the localization of the absorption bands is independent of the oxygen concentration testifies to the unchanged nature of the color centers. The intensities of the absorption bands suggest a difference in density between the point defects. Until proven otherwise, the point defects can be trap charge carriers and therefore affect the electrical resistivity of crystals and can induce their coloration.

3.4. Electrical resistivity characterization

In Table 2, the electrical resistivity measurements demonstrate that the presence of oxygen in the growth atmosphere decrease the electrical
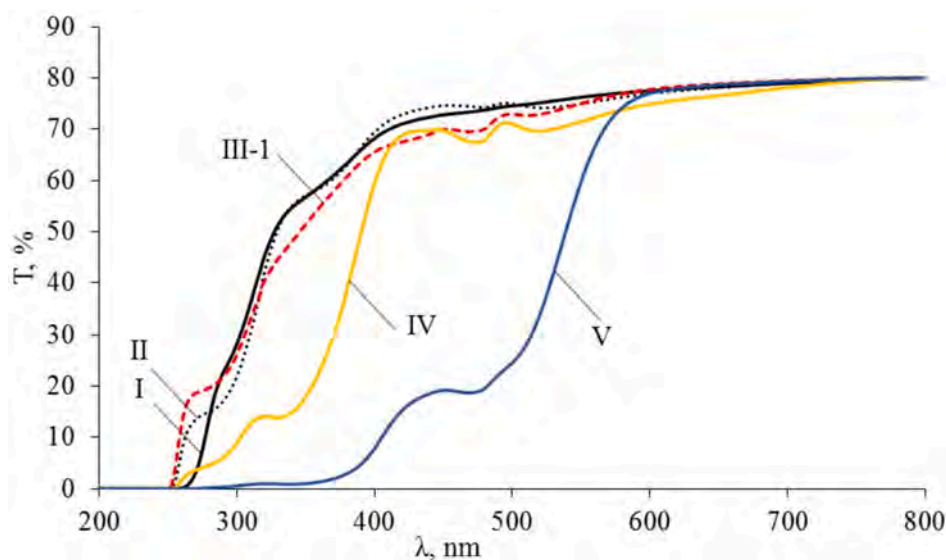
Fig. 7. UV–visible transmission spectra of LGT crystal as a function of growth atmosphere.

resistivity of the LGT crystals. The LGT grown under Ar was practically twice as resistant as LGT crystallized under Ar + 1% O$_2$. By comparing LGT crystals as a function of the growth atmosphere, it turns out that the addition of a small amount of oxygen (Ar + 0.1%) decrease the electrical resistivity. Quite often, at room temperature, the resistivity of piezo-electric materials is quite high and belongs to the range (10^{13}–10^{15} Ω/cm) [37]. It strongly depend on the structural perfection of the crystal. Therefore, it is important to control the composition during the growth process and limit the composition evolution such Ga2O3 losses and oxygen vacancies. Such structure defects deteriorate the crystals properties.

4. Conclusions

High quality langatate (LGT) single crystals of diameter 50 mm were grown by Czochralski technique using iridium crucible as container and argon mixture (0.1–1%O$_2$) atmosphere. Under stationary stable growth conditions, highly transparent, orange LGT single crystals were crystallized without defects such cracks, bubbles, grains boundary and inclusions. The crystals grown in argon atmosphere were colorless and the addition of 1–2% excess Ga$_2$O$_3$ allowed the obtainment of optically perfect crystals. Along Z-axis, the LGT grown crystals from the melt by Czochralski technique have tendency to be facetted, which has an important effect on the external crystal appearance. The contrast coloration evolution of the grown crystals strongly depend on the oxygen content in the chamber. The transmission spectra of the grown crystals showed the relationships between LGT coloration and oxygen vacancies. The oxygen content in the gas atmosphere decrease the electrical resistivity and the LGT crystal grown in inert pure argon environment has the best electrical resistivity.

CRediT authorship contribution statement

B. Boutahraoui: Crystal growth of LGT by Czochralski technique in France. **E.A. Ghezal:** developing automatic LGT Czochralski crystal growth. **A. Nehari:** Optical characterization and transmission results interpretation. **K. Zaidat:** Chemical LGT composition analysis to confirm the stoichiometric LGT composition. **M. Allani:** electrical resistivity measurement. **H. Cabane:** Cutting and polishing LGT Crystal, crystal orientation. **M. Dumortier:** LGT Z-Seed preparation Cutting and polishing LGT Crystal. **I. Gerasymov:** LGT crystal growth by Czochralski technique in Ukraine. **O. Sidletskiy:** discussion and exchange on the results and paper contribution writing. **S. Obbade:** Solid state reaction,

Table 2
Electrical resistivity LGT crystals as a function of the growth atmosphere.

Growth atmosphere	Ar	Ar+0.1% O$_2$	Ar+0.3% O$_2$	Ar+0.5% O$_2$	Ar+1% O$_2$
Resistivity, ρ (10^{14} Ω cm)	97.45	89.37	81.38	70.33	53.37

phases characterization and Ga2O3 oxide optimization in the melt, S. Jouffret: Phases characterization and Ga2O3 oxide optimization in the melt. **K. Lebbou:** Results interpretation and writing paper.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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