

## PRELIMINARY RESULTS ON CRYSTAL GROWTH OF BULK $\text{In}_x\text{Ga}_{1-x}\text{As}$ USING TLZ METHOD

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### ABSTRACT

Preliminary growth experiments have been carried out to investigate the possibility of growing bulk  $\text{In}_x\text{Ga}_{1-x}\text{As}$  crystals with homogeneous composition using the travelling liquidus zone growth method. The experimental environments were made by optimizing several growth conditions for the growth of  $\text{In}_x\text{Ga}_{1-x}\text{As}$  crystals with 10 mm in diameter. The experiments were performed for two different sets of growth parameters. Although single crystal was not obtained from the preliminary experiments, crystals with a homogeneous compositional profile ( $x \sim 0.3$ ), near the seed-crystal interface, extending up to the length of few millimetres were obtained.

### 1. INTRODUCTION

Ternary bulk  $\text{In}_{0.3}\text{Ga}_{0.7}\text{As}$  mixed crystal is an attractive lattice-matched substrate material for InGaAs-based laser diodes operating at 1.3  $\mu\text{m}$  wavelength in the next generation of optical communication systems. The lattice parameters in this substrate material can be tuned precisely by changing the composition. In order to fabricate a wide variety of devices on an  $\text{In}_{0.3}\text{Ga}_{0.7}\text{As}$  substrate, it is a basic requirement to have a high-quality large size  $\text{In}_{0.3}\text{Ga}_{0.7}\text{As}$  single crystal with homogeneous composition. However, it is difficult to grow such crystals, because convection in a melt combined with segregation at the time of solidification, causes compositional variation as well as local polycrystallization in the grown crystals. Particularly, it is more difficult to control the compositional homogeneity near the composition 0.3, in which the separation between the solidus and

liquidus lines is far apart in the phase diagram of  $\text{In}_x\text{Ga}_{1-x}\text{As}$  system.

Several growth methods, such as liquid encapsulated Czochralski [1], Bridgman [2], vertical gradient freeze [3], and multi-component zone melting (MCZM) [4], have been applied to grow bulk  $\text{In}_x\text{Ga}_{1-x}\text{As}$  crystals. However, a large size of single crystals with homogeneous composition at  $x = 0.3$  could not be obtained. Although Kodama et al. were succeeded to grow  $\text{In}_{0.3}\text{Ga}_{0.7}\text{As}$  single crystal having 15 mm in diameter using the modified MCZM method [4], the crystal length was limited to only 4 mm due to polycrystallization and breakage.

It is believed that the stable transportation of solute is fundamental to homogeneous crystal growth. On the basis of this, Kinoshita et al. [5] have developed a new crystal growth method, named as, traveling liquidus zone (TLZ) method, which is a modified form of the vertical Bridgman method. In this method, crystal growth is done by forming a narrow liquidus zone with the combination of the temperature gradient in the furnace and the compositional gradient in the feed material. Therefore, it is required to characterize the compositional profile of the feed material for optimizing the TLZ growth conditions. This method has already shown some successes in growing compositionally homogeneous single crystal with 20 mm length. However, the diameter of the crystals was limited to only 2 mm, which is too small for its practical use as a substrate. In the present study, the TLZ growth experiments were performed in order to grow compositionally homogeneous bulk  $\text{In}_x\text{Ga}_{1-x}\text{As}$  crystal with 10 mm in diameter and the results

obtained from the preliminary experiments have been presented.

## 2. EXPERIMENTAL PROCEDURE

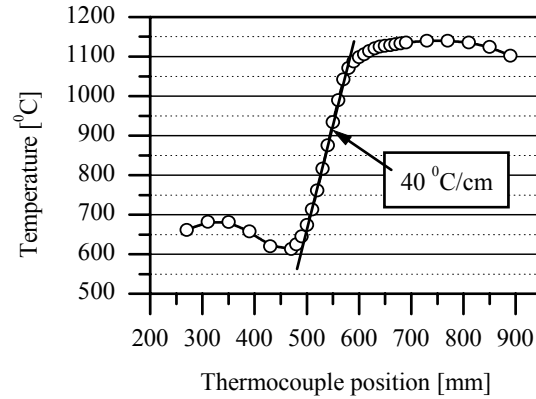
### 2.1 Preparation of Feed Material for TLZ Growth Method

Several  $\text{In}_x\text{Ga}_{1-x}\text{As}$  polycrystalline source materials with 150 mm in length and 32 mm in diameter were prepared using the directional solidification method. These source materials were found to have a gradient in compositional profile, ranging from  $x = 0.98$  at one end to about  $x = 0.02$  at other end. The compositional profile was determined in a nondestructive way by using Raman scattering (RS) technique [6]. The feed materials were prepared from the source material in the shapes of cylinders with 10 mm in diameter and 100 mm in length. These feed materials were cut from a position of the source material so that the composition in the feed material had a gradient from  $x = 0.9$  to about  $x = 0.1$ . In order to check the suitability of these feed materials for the TLZ growth technique, the compositional profiles of these feed materials were measured once again using RS, after they were cut from the source material.

### 2.2 Preparation of Experimental Conditions for TLZ Growth Method

#### 2.2.1 Temperature profile measurements

In order to decide the set values of the heaters as well as the position of cooling plate in the furnace for obtaining the desired temperature gradient for TLZ growth, the temperature profile was measured by installing one thermocouple at the centre of the furnace. This thermocouple was moved along the

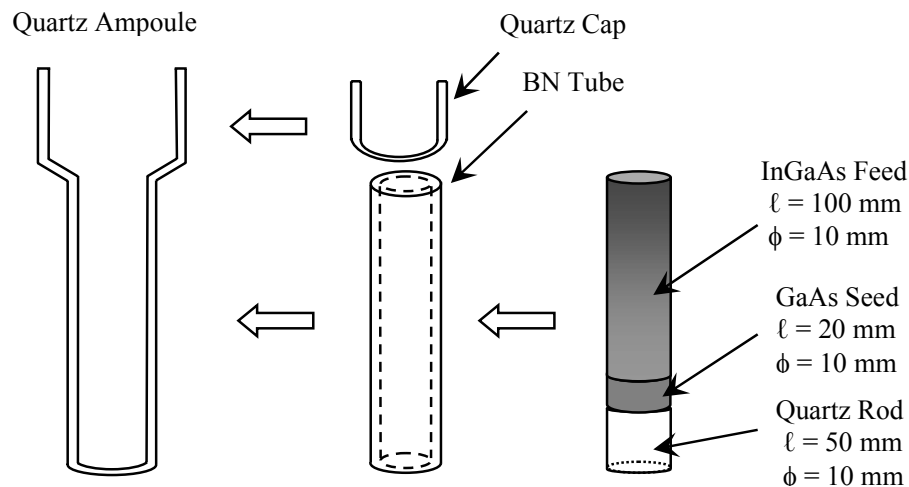


**Fig. 1** Temperature profile used for the TLZ growth process.

length of the furnace, and in this way temperatures at close points inside the furnace were measured. A large number of experiments were performed to obtain the desired temperature profile suitable for the TLZ growth. Figure 1 shows the temperature profile, which was used for the growth process in the present experiments.

#### 2.2.2 Design for growth process

A schematic of the design for the growth process is shown in Fig. 2. GaAs single crystal with 10 mm in diameter and 20 mm in length was used as the seed. Facing the InAs-rich side of the feed material towards the seed, the seed and the feed material were put inside a boron nitride (BN) tube of 10 mm in inner diameter and 170 mm in length. In order to provide a heat sink to the seed, the remaining 50 mm of the BN tube next to the seed was filled with a quartz rod of 10 mm in diameter and 50 mm in length. This quartz rod was mirror-polished at both



**Fig. 2** A schematic of the seed and feed arrangement in the quartz ampoule for the crystal growth.

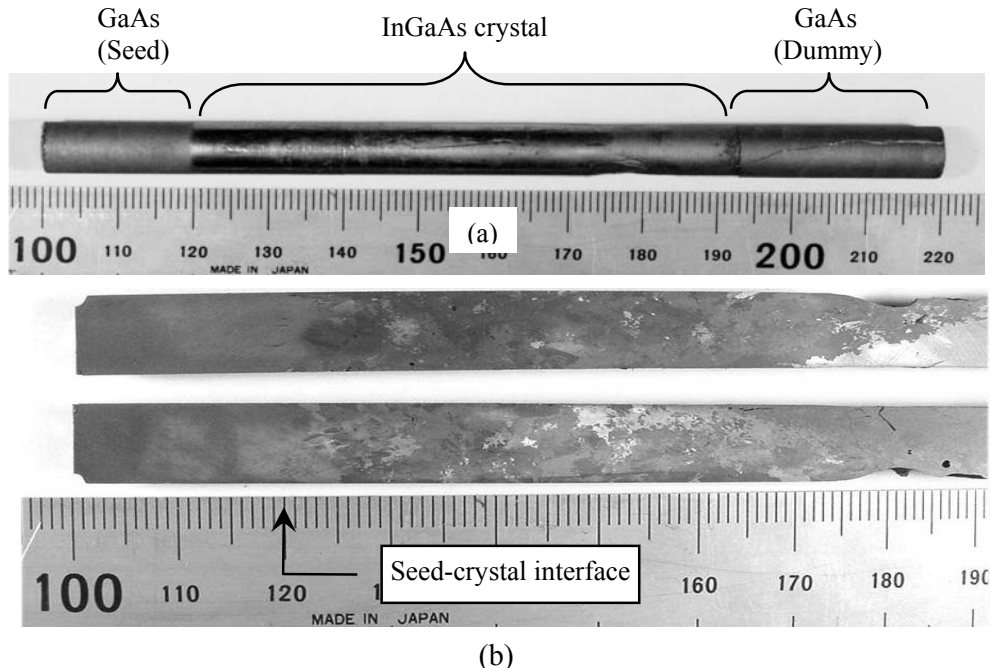
ends, in order to facilitate a better radiation. The BN tube, along with the feed, seed and quartz rod, was put inside a flat-bottom quartz ampoule, which was then sealed under vacuum using a quartz cap. A special quartz stage was designed to hold this ampoule, which was suitable for installing eight thermocouples at various positions, and for heat radiation at the bottom. Five thermocouples were installed outside the stage, whereas three thermocouples were installed inside the stage. In this way, it was possible to measure the temperature in the vicinity of seed-crystal interface, which is necessary to determine the melt-zone in the TLZ growth method.

### 3. GROWTH RESULTS

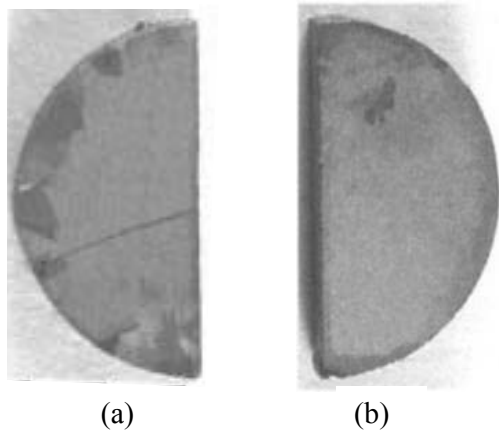
Using the above mentioned growth design; TLZ growth were carried out under the two different experimental conditions. In the first growth experiment, instead of 100 mm, the feed material of 72 mm in length was used. Rest space of 28 mm was filled with a dummy GaAs crystal. At the starting position, the seed-crystal interface was kept at 1024 °C, where it was soaked for one hour, and then it was translated at a speed of 0.5 mm/hr. After a total growth of 100 hrs, the crystal was cooled down at a slow cooling rate of 25 °C/hr. After growth, the crystal was sliced along growth length into two half-cylinders, so as to investigate the inner part of the

crystal. Pictures of this crystal before and after slicing are shown in Fig. 3. A careful observation of the picture in Fig. 3(b) reveals that the center of the seed-crystal interface has a large crystal grain, where as the outer part of the crystal at the seed-crystal interface has small grains. This indicates that there was a single crystal growth for an extent of about 2 mm at the center of the seed-crystal interface. In order to investigate this fact, a 1 mm thick wafer was sliced from one of the half-cylinders, near the seed-crystal interface, in the direction perpendicular to the growth, so that the seed-crystal interface lies within the wafer. Figures 4(a) and 4(b) show the two faces of this wafer, 4(a) is from the crystal side and 4(b) is from the seed side. As expected, Fig. 4(b) shows a single crystal, because it is seed. It becomes clear from Fig. 4(a) that the outer side of the grown crystal has small grains, whereas the central part shows large grain size, indicating a single crystal growth at the center. However, this single crystal growth could not extend more than 2 mm in length.

The other half-cylindrical part of the grown sample was characterized for its compositional profile using photoluminescence (PL). The results are shown in Fig. 5. The composition was found to be almost constant near a value of  $x = 0.3$ , for a length of about 3 mm close to the seed-crystal interface. After a length of 3 mm, the composition was suddenly increased to a large value. In the first growth experiment, it was possible to grow a crystal



**Fig. 3** (a) Picture of as-grown crystal in the first growth experiment. (b) Cross-sectional cut along the growth axis showing the seed-crystal interface.



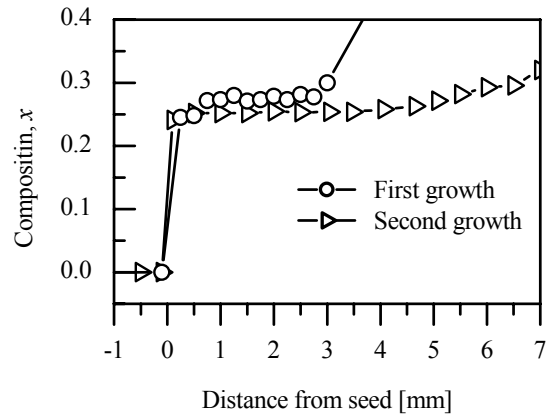
**Fig. 4:** Pictures of both faces of a 1 mm thick wafer sliced at the seed-crystal interface in which interface lies within the wafer. Picture (a) is from the crystal side and (b) is from the seed side.

with constant composition of about  $x = 0.3$  for a length of about 3 mm, and also to get large grain size for a growth length of about 2 mm.

In order to achieve a longer length of constant composition, another growth experiment was performed with slightly modified growth parameters. In this case, the soaking time was increased from 1 hr to 2 hrs and the translational speed was changed from a constant value of 0.5 mm/hr to 0.3 mm/hr for the first 5 mm of growth and then 0.8 mm/hr for rest of the growth. Large grain size could not be obtained by this growth, as it was obtained in the previous growth. However, a better compositional homogeneity for a growth length of about 5 mm was obtained. A comparison of compositional profile measured by PL near the seed-crystal interface for the first and the second growth experiments is also shown in Fig. 5. In the first growth experiment, better crystallinity was obtained than the second growth experiments. However, better compositional homogeneity for a longer growth length was obtained in the second growth experiment.

#### 4. CONCLUSIONS

Using the TLZ growth technique, the possibility of growing compositionally homogeneous bulk  $\text{In}_x\text{Ga}_{1-x}\text{As}$  crystals with 10 mm in diameter has been investigated. The experimental environments were made by optimizing several growth conditions and then the growth experiments were performed in two different conditions. In the first experiment, single crystal growth was observed at the central part of the ingot near the seed-crystal interface; however, polycrystallization was started from the periphery of



**Fig. 5** Comparison of compositional profiles for the first and the second growth experiments

the ingot. In this growth, the compositional homogeneity of the crystal was found to be approximately  $x \sim 0.3$  extending to about 3 mm in length from the seed-crystal interface. In the second growth experiment, although the crystalline quality was found to be poor than that of the first growth, better compositional homogeneity was obtained for a length of about 5 mm from the seed-crystal interface.

#### REFERENCES

- [1] K. Nakajima and T. Kusunoki, "Constant temperature LEC growth of InGaAs ternary bulk crystals using the double crucible method," *J. Cryst. Growth*, vol. 169, pp. 217-222, April 1996.
- [2] K. Nakajima, T. Kusunoki, and K. Otsubo, "Bridgman growth of compositionally graded  $\text{In}_x\text{Ga}_{1-x}\text{As}$  single crystals for use as seeds for  $\text{In}_{0.25}\text{Ga}_{0.75}\text{As}$  crystal growth," *J. Cryst. Growth*, vol. 173, pp. 42-50, August 1996.
- [3] Y. Nakajima, K. Nakajima, K. Otsubo, and H. Ishikawa, "InGaAs single crystal using a GaAs seed grown with the vertical gradient freeze technique," *J. Cryst. Growth*, vol. 197, pp. 769-776, August 1999.
- [4] S. Kodama, Y. Furumura, K. Kinoshita, H. Kato, and S. Yoda, "Single crystalline bulk growth of  $\text{In}_{0.3}\text{Ga}_{0.7}\text{As}$  on GaAs seed using the multi-component zone melting method," *J. Cryst. Growth*, vol. 208, pp. 165-170, July 2000.
- [5] K. Kinoshita, H. Kato, M. Iwai, T. Tsuru, Y. Muramatsu, and S. Yoda, "Homogeneous  $\text{In}_{0.3}\text{Ga}_{0.7}\text{As}$  crystal growth by the traveling liquidus-zone method," *J. Cryst. Growth*, vol. 225, pp. 59-66, February 2001.
- [6] M. R. Islam, P. Verma, M. Yamada, M. Tatsumi, and K. Kinoshita, "Micro-Raman characterization of starting material for traveling liquidus zone growth method," *Jpn. J. Appl. Phys.*, vol. 41, pp. 991-995, August 2001.