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Growth of Bulk GaN Single Crystals by the Pressure-Controlled Solution Growth Method

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1. Introduction

GaN based optical devices such as blue-light LEDs and lasers have been developed using sapphire substrates [1,2]. For these devices, it is however desired to use GaN substrates since the device production yield can be increased by using conductive GaN substrates instead of insulating sapphire substrates. GaN substrates also have advantages in having cleaved faces for laser mirror fabrication.

Since GaN has a high melting point (>2500°C) and an extremely high dissociation pressure, it was difficult to grow single crystals. Recently, a high pressure solution growth (SG) method was applied to the growth of GaN bulk crystals. In the present work, we have first developed a pressure-controlled high pressure solution growth (PC-SG) method and applied it to the growth of GaN bulk crystals.

2. Experimental Procedure

In the conventional solution growth, the solvent is saturated by solutes and the temperature is decreased in order to realize the supersaturation for growing crystals. This process has however following disadvantages; (1) constitutional supercooling easily occurs, (2) the growth rate decreases, (3) the large part of the solvent is left, as can be predicted from Fig. 1. Because of these disadvantages, it is believed that solution growth is not beneficial in preparation of compound semiconductor materials. In order to overcome these disadvantages, we have first applied the PC-SG method. In this method, the solute gas pressure is continuously applied during crystal growth and the supersaturation is realized by applying an over pressure without any temperature decrease. This method has therefore following advantages; (1) constitutional supercooling does not occur, (2) the growth rate is constant, (3) all solvent can be principally converted to crystals, as can be seen from Fig. 1. This pressure-controlled solution growth can be easily performed only when one constituent of the compound is in the gas phase at room temperature. The case of GaN is very appropriate to apply this method.

In the present work, we have made a preliminary study for examining the validity of the PC-SG method by using a high pressure furnace (Fig. 2). About 400g 6N-Ga was charged in a pBN crucible and was set in the furnace. After purging the furnace by nitrogen gas, the furnace was heated with pressurizing by nitrogen. Crystal growth was performed at 1350-1600°C under the nitrogen gas pressure 7000-10000 kgf/cm^2. For holding the desired nitrogen pressure during crystal growth, the pressurizing piston attached to the high pressure furnace was continuously pressed.

3. Results and Discussion

Fig. 3 shows a GaN single crystal grown at 1450°C for 9 hours under 10000 kgf/cm^2. The crystal size was about 7 mm square and the shape was hexagonal. It was found by X-ray diffraction that the normal axis of the crystal platelet was the (0002) plane, and was found to be c-axis oriented. The crystallinity was examined by plain-view TEM and cross sectional TEM, and any defects could not be observed (Fig. 4). The dislocation density was thus predicted to be less than 10^6 cm^{-2}. By photoluminescence measurement at 4.2 K, it was found that the intensity of the edge-emission at 3.5 eV was twice as that of the yellowish emission at 2.3 eV (Fig. 5). It was thus found that crystals grown by the present method were of good crystallinity.

4. Conclusions

We have made a preliminary work to examine the validity of the PC-SG method and found that GaN single crystals can be obtained only by holding the Ga solution at a certain temperature under an over pressure without decreasing the temperature itself. This result was very encouraging to overcome the disadvantages of the solution growth method. The present work may open a way to convert all Ga solvent to GaN crystals.

Acknowledgments

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References

[1] S. Nakamura, T. Mukai and M. Senoh: Appl. Phys. Lett. **64** (1994)1687.

[2] S. Nakamura, M. Senoh, S. Nagahama, N. Iwasa, T. Yamada, T. Matsushita, H. Kiyoku and Y. Sugimoto: Jpn. J. Appl. Phys.**35**(1996)L74.

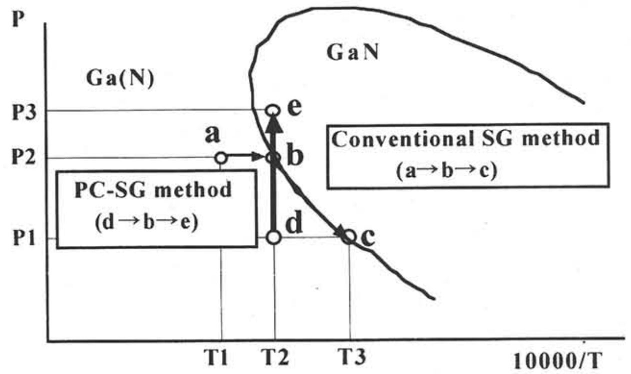

Fig. 1 Schematic P-T diagram of GaN.

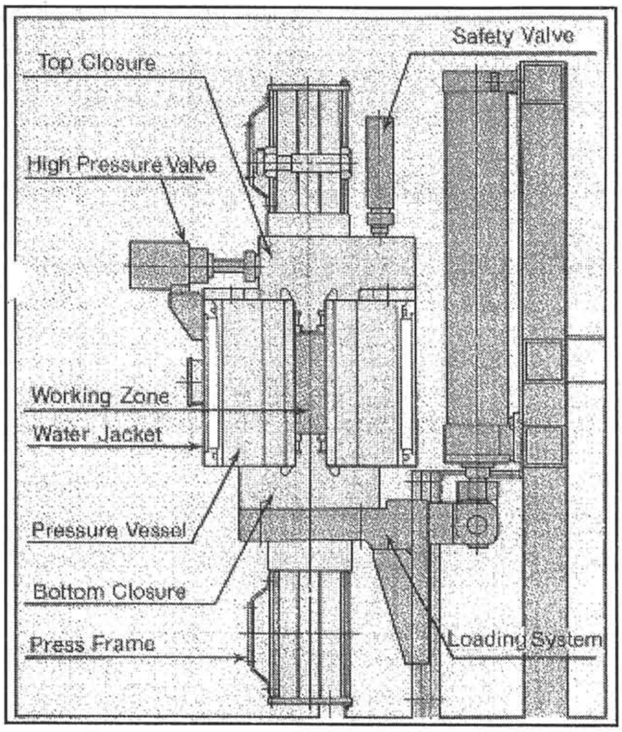

Fig. 2 Schematic cross sectional view of the high pressure furnace.

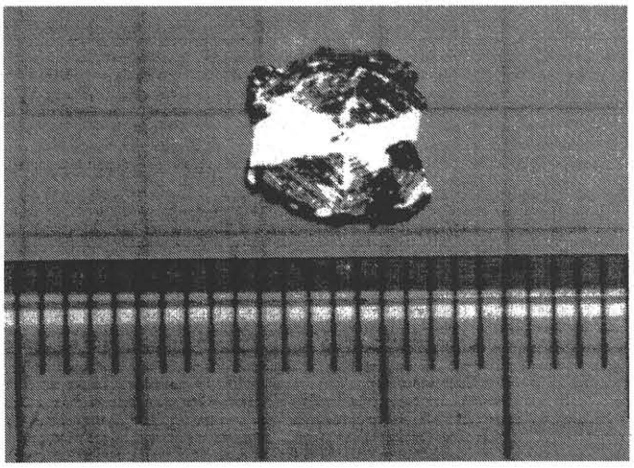

Fig. 3 GaN single crystal grown by the pressure-controlled high pressure solution growth (PC-SG) method.

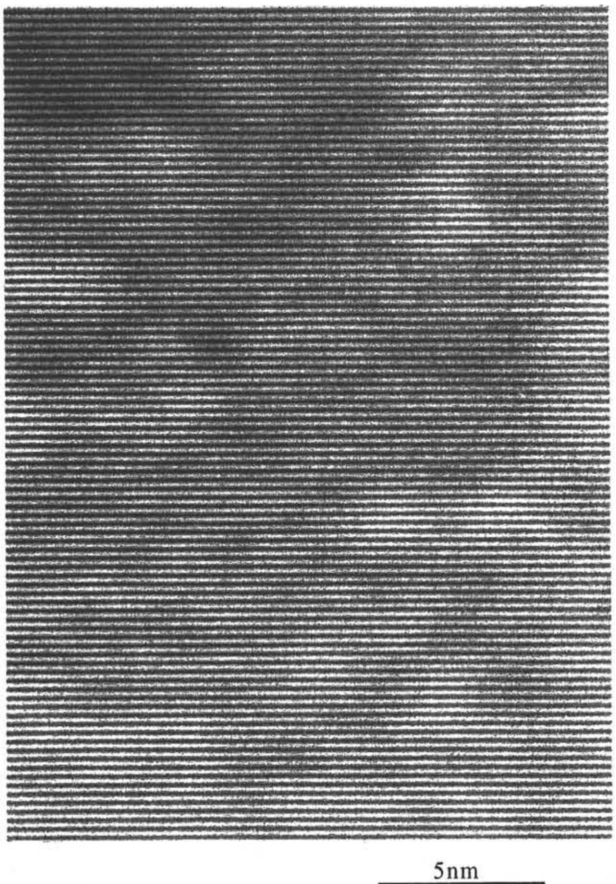

Fig. 4 Cross sectional TEM image of the GaN single crystal.

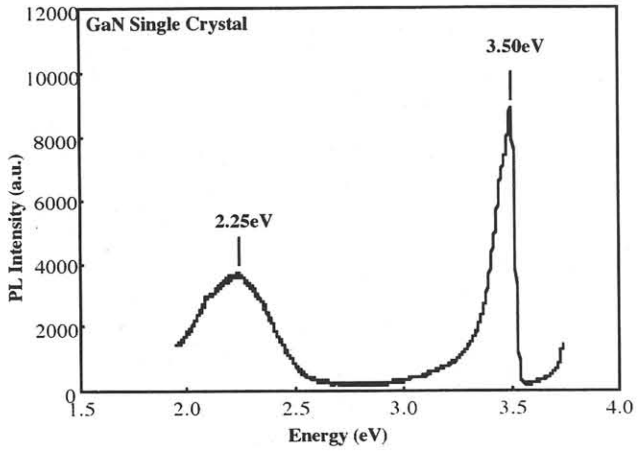

Fig. 5 Photoluminescence spectrum at 4.2 K of the GaN single crystal.