Growth of bulk GaN Crystal by Na Flux Method

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Abstract
With current technologies, dislocation-free bulk shape GaN crystals with centimeter-size can be grown from small seed crystals. To enlarge the diameter of bulk shape GaN crystals, we have developed the coalescence of GaN crystals from many isolated small seeds.

1. Introduction
Na flux method developed by Yamane can grow GaN crystals in a Ga-Na mixed solution at relatively low pressure of nitrogen atmosphere (<5MPa) and at temperature range of 750~900 deg.C. At the beginning, we have utilized the seed crystal fabricated by the vapor phase growth method in order to make a large diameter GaN crystal. In spite of the poor quality seed substrate with high dislocation density, high quality GaN crystal with the dislocation density around 10^3/cm^2 could be obtained. It is possible to grow a 4-inch GaN crystal on a HVPE-GaN substrate. However, there should be the limitation of the quality and size of GaN crystal grown on the HVPE substrate containing the residual stress.

Recently, we have developed two new techniques to grow large dislocation-free GaN by Na flux method.

2. Growth of GaN crystals from small GaN seeds
First one is Point Seed (PS) technique\(^1\). This technique can be realized by putting a sapphire plate with a small hole (0.5 – 1.5 mm in diameter) on a GaN plate seed. Centimeter-sized bulk GaN single crystals with large dislocation-free areas could be fabricated by this technique. Cathodoluminescence measurement at the interface between the seed and the grown crystal has revealed that almost all dislocations propagated from the GaN seed were bent and terminated at the initial growth stage.

3. Coalescence growth of GaN crystals by Na flux method
Second one is coalescence growth of multi-GaN crystals in order to fabricate a large diameter single GaN crystal within a short period\(^2\). As a first step, we grew two GaN point seeds and coalesced them. Two GaN point seeds were established by mounting a sapphire plate with two small holes. The coalescence direction was a-direction. Other experimental conditions were same as above. We have found the two GaN crystals grown from two separate seed area coalesced without generating dislocations at a coalescence boundary. X-ray rocking curve measurements revealed that miss-orientation of c-axis of two crystals around a coalescence boundary gradually diminished during the growth. The grown GaN crystal can remove from substrate easily during the growth. This phenomenon is effective to reduce the stress in the grown GaN crystal.

Fig. 1 Photograph of the GaN crystals grown from Ba-doped solution on the small GaN seed crystal.

Fig. 2 Schematic of coalescence growth from two isolated small seed crystals and SEM images of the sliced crystals coalescing along the a-direction.

The size of the GaN crystal can be increased as increasing the number of seed crystals. Up to now, we succeeded to fabricate 2-inch GaN crystals by the coalescence technique. Some of the crystals have very large curvature radius (~100 m), which exceed the detection limit of a Rigaku SmartLab X-ray diffractometer. Additionally, the point contact Schottky diode at the coalescence boundary of the crystal could work normally.
4. Conclusions
In this paper, we reported the recent advances in the growth of GaN crystals by Na flux method. In the case of the growth on a small GaN seed, Dentimeter-sized buld GaN crystal could be obtained. No dislocations could by observed by the CL measurement from the GaN crystal grown on the small seed crystal. It is possible to coalesce GaN crystals grown from isolated small seed crystals without generation dislocations at boundary. The coalescence growth of GaN crystal from periodical arranged many small seed crystals seems to become a key technique for fabricating large-diameter high quality GaN crystals.

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References
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