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Structural characterizations of GaN nanowires and GaInN/GaN multi-quantum shells grown by MOVPE

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Abstract

Structural characterizations of core-shell type nanowire-based materials are difficult because of their tiny 3D structure. In this paper, we prepared the GaInN/GaN multi quantum shell (MQS) grown on the sidewall of the n-GaN nanowire, and scanning transmission micro-scope (STEM) and 3D atom probe are employed for structural characterization of such samples. As a result of 3D atom probe measurement, an out-diffused profile of In, where In atoms move from the wells to the outer barriers, was observed. This is considered that the incorporation of In in the outer barriers come from In–droplets caused on the top c-plane during the growth of GaInN.

1. Introduction

Recently, GaN nanowires and its application to light-emitting devices attract much attention [1]. Nanowire discussed in this paper is a hexagonal columnar crystal grown in the vertical direction from the substrate surface. Its diameter is less than 1 μm, and the height is from several hundred nm to several μm.

A structure composed of the GaN nanowire and the multi-quantum shell (MQS) active region, which is recognized as “core-shell structure”, has several advantages, such as basically dislocation-free material [2], a polarization-free face on the sidewall of the nanowires [3], and a flexibility of active area by changing the height of nanowire. Therefore, the MQS/nanowire structure has a potential to solve some problems in current LEDs. For instance, the efficiency droop in LEDs [4], which has been an insoluble problem for a long time, could be suppressed by increasing the volume of active region with a large length of nanowire or thick MQS. According to the theoretical calculation, the arrangement of MQS could provide a higher optical confinement factor in laser cavity [5], when it is applied to semiconductor lasers. Therefore, the structure consisting of GaN nanowires and MQS active regions would give the high performance LED and lasers in the near future. To realize such devices, high quality nanowire-based material has to be grown. Thus, in this paper, the structural characterizations of GaN nanowires and GaInN/GaN MQS grown by MOVPE are presented.

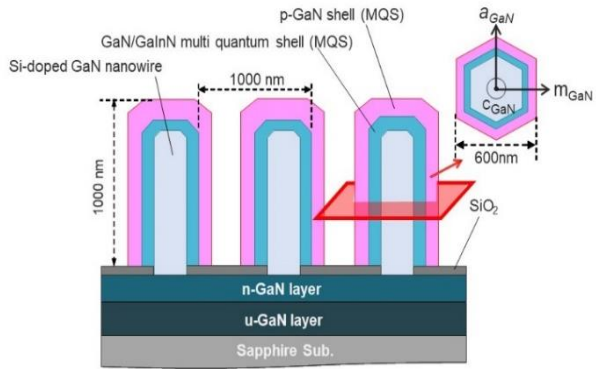

Figure 1 Schematic of GaN nanowires and GaInN/GaN MQS.

2. Experimental method and Results

Figure 1 shows a schematic diagram of a n-GaN nanowire and GaInN/GaN MQS covered by a p-GaN shell. The n-GaN template was prepared by the MOVPE growth technique on a c-plane sapphire substrate. A SiO_2 film was deposited on the n-GaN by a magnetron radio-frequency sputtering technique. The thickness of SiO_2 was approximately 30 nm. Then, nanoimprinting lithography and the inductive coupled plasma etching was carried out to form a triangular arrangement of openings of SiO_2 mask patterns with a diameter of 250 nm and pitch of 1,000 nm.

The Si-doped n-GaN nanowires were grown on n-GaN through the openings of the SiO_2 film by the continuous mode of MOVPE. TMGa, NH_3, and SiH_4 were used as precursors with a very low V/III ratio of 20. The growth temperature and reactor pressure were maintained at 1120 °C and 90 kPa, respectively. After the n-GaN nanowires, the MQS structures were successively grown around

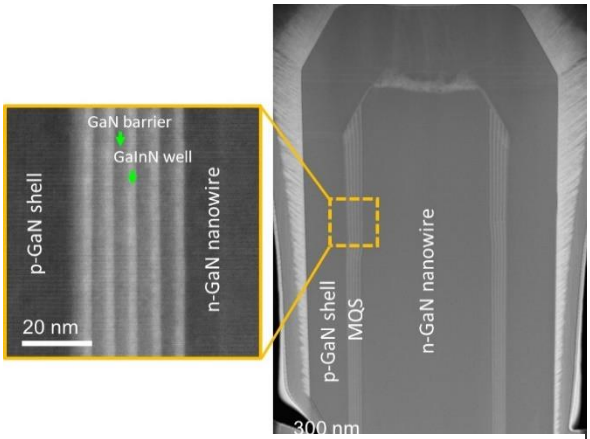

Figure 2. Cross-sectional HAADF-STEM images in MQS/nanowire.

the n-GaN nanowires adding TMIn with a V/III ratio of 4000. The growth temperature and reactor pressure for the MQS were 740 °C and 90 kPa, respectively.

The detailed structure and compositional profile of the GaInN/GaN MQS structures were investigated by scanning transmission electron microscope (STEM) and 3D atom probe tomography. Figure 2 shows the high-angle annular dark field scanning transition electron microscopic (HAADF-STEM) images of cross-sectional MQS. The GaInN quantum-shells have a brighter contrast in the STEM image, and the n-GaN nanowire looks defect-free in the MQS on m-plane. There are no stacking faults extended from the interface between the n-GaN nanowire and MQS, which are frequently observed in this kind of structure. The MQS on the semi-polar plane cannot be seen clearly, because of the low growth rate. Especially the MQS of top c-plane area looks quite rough. This may be due to the rough surface of n-GaN nanowire after cooling down duration for the MQS growth, and the MQS might cause decomposed Ga and In droplets. The film thickness of the GaN barriers and GaInN wells were approximately 2.3 nm and 4.2 nm, respectively.

Figure 3 shows the In fraction line profile of MQS obtained by the 3D atom probe measurement. The In fraction in the GaInN wells in the MQS is ranging from 0.17 to 0.20.

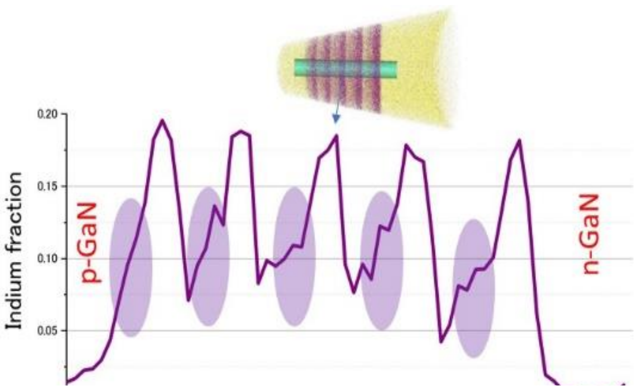

Figure 3. 3D atom probe tomography (upper inset) and extracted line profile of indium in GaInN/GaN MQS

And it seems to show out-diffusion of In (indicating grey ovals). We also confirmed that the opposite side (right – hand side of the nanowire) has the reversed In profile. Such the out-diffusion of In in GaInN/GaN MQW has not been reported, to the authors best knowledge. The most likely cause is thought to be the In vaporization from the In droplets on the top area of the nanowire. We actually observed the In droplets in the samples without the p-GaN shell growth. To suppress the In droplet formation, it is necessary to modify the growth sequence of MOVPE.

3. Conclusions

The structure composed of MQS and n-GaN nanowire grown by MOVPE shows basically high crystalline quality and uniform MQS composition and thickness on the m-plane, excepting the rough surface of top c-plane and the out-diffusion of In profile in the MQS. From the In line profile obtained in 3D atom-probe tomography, out-diffusion of In was observed. It might be caused by In vaporization from the In droplets on the c-plane. It should be suppressed by the modification of the growth condition.

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