P-13-9

Growth of vertical GaN Nano-Column on Au Droplet/Si(111) Substrate using Pulsed Flow MOCVD Method

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1. Introduction

GaN and its related alloys are generally useful to optical devices such as light emitting diodes and photodetectors in blue and ultraviolet regions because of their wide direct band gap and high electrical and thermal conductivity. However, there is still a great need for improvement of the light-extraction efficiency and internal quantum efficiency. Many nanotechnologies have been implemented to improve the efficiency GaN layers were studied in order to create building blocks of various sizes and shapes. Therefore, the development of new methodologies to improve the existing growth techniques is very important for realizing low-dimensional nanostructures. GaN nano-columns are useful to improve the performance of low-dimensional high power light sources because it is anticipated that high optical gains and low lasing thresholds will be achievable when the nano-column diameter is smaller than the excition radius [1]. Recently, GaN nano-column preparation is typically based on the vapor-liquid-solid (VLS) growth mechanism and involves the use of catalysts such as Au, Ni, Fe, or In [2~4]. The catalyst creates a highly selective growth environment with growth occurring only at the catalyst location. GaN nano-columns using metallic catalyst can be integrated on silicon substrates for use as III-nitride nano-column emitters and as silicon based nano photo-detectors for data communications by photons. The major challenge faced by the GaN nano-column grown by metalorganic chemical vapor deposition (MOCVD) is that the directions of nano-columns are disordered. In order to grow GaN nano-column in unidirectional, we have adapted two step methods where Au droplets are annealed prior to nano-column growth to obtain uniform distribution and flow of the precursors into the chamber was by pulse method to obtain unidirectional growth. In this paper, we have studied the growth of GaN nano-column using pulse flow method by MOCVD technique.

2. Experimental procedure

GaN nano-column structures were grown on gold (Au) coated Si(111) oriented substrates by MOCVD using a horizontal quartz reactor. The reactant precursors were trimethlygallium (TMGa) and ammonia (NH$_3$) for Ga and N, respectively. Hydrogen (H$_2$) was used as carrier gas. Prior to nano-column growth, the Si substrate surface was treated using a conventional cleaning procedure and a gold thin film was deposited using an ion sputtering system. The Au coated Si substrate surface was cleaned in acetone and isopropyl alcohol to remove any contamination on the thin film surface. The Au coated Si substrate was annealed at 800 $^{\circ}$C under hydrogen ambient for 5 min. The group III and group V precursors were introduced alternately in the sequence: TMGa, 1 sccm, 20 s; NH$_3$, 1 slm, 60 s in a pulse method for three cycles to form the seed for the nano-column growth. Further, GaN growth was began at 1040 $^{\circ}$C for 30 min with an ambient pressure of 150 torr.

The surface morphology and structural characteristic were analyzed using scanning electron microscopy (SEM) and field emission SEM (FE-SEM).

3. Results and discussion

The GaN nano-columns were grown on Au-coated Si(111) substrates by flowing TMGa in a NH$_3$ and H$_2$ atmosphere at approximately 1040 $^{\circ}$C for 30 min. The self-assembled technique involves four major steps: (i) Ion coating of the Au thin film on the Si(111) using an ion coating system, (ii) thickness control of the gold thin film, (iii) the formation of GaN seeds on Au droplet by pulsed MOCVD, and (iv) the growth of the GaN nano-columns.
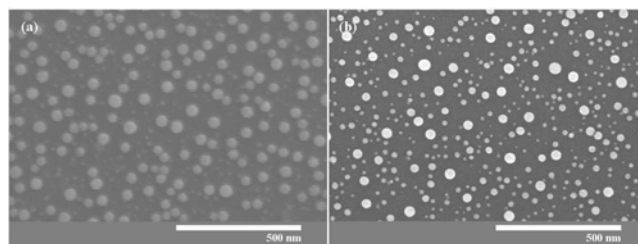

Fig. 1. FE-SEM images of Au droplets for two different Au deposition thicknesses (a) 10 nm and (b) 15 nm after annealing at 800 $^{\circ}$C for 5 min.

The first step for the growth GaN nano-column is the condensation and adhesion of Au atoms onto the surface of the Si(111) substrate to form nano sized catalytic islands.
Fig. 1 shows the SEM images of the Au/Si template after annealing at 800 $^{\circ}$C for 5 min for the two different Au deposition thicknesses 10 nm and 15 nm. Fig. 1(a) shows a uniform Au cluster distribution on the silicon surface of size ∼50 nm. Whereas, for 15 nm gold thickness, the clusters size varied from 10 nm ∼ 60 nm unevenly distributed.

The critical feature of this mechanism is the presence of intermediates formed by the precursors and nano metallic islands, which act as catalysts between the vapor feed and solid growth at an elevated temperature and Au thin film
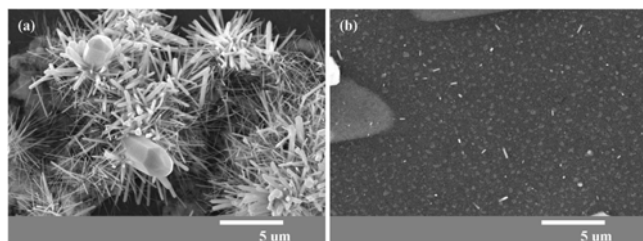
thickness in the growth process.

Fig. 2. SEM images of GaN nano-columns (a) without Au annealing and (b) with Au annealing.

Fig. 2(a) shows the SEM image of nano-columns without using Au annealing. The bundles of nano-columns were grown at the few regions where Au catalysts were agglomerated. GaN nano-columns could be grown all over the region using Au annealing to make droplet as shown in Fig. 2(b). However, the directions of nano-columns were disorders [5]. The direction of nano-column is very important to use for devices.
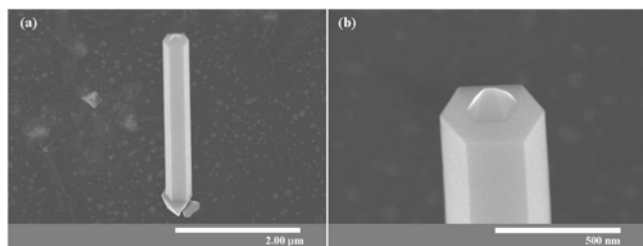

Fig. 3. FE-SEM images of vertical nano-columns using pulsed flow MOCVD.

In order to improve the unidirectional growth, pulse flow MOCVD was used to make the GaN seeds/Au droplet. By using this method, we can grow the vertical GaN nano-columns. Figure 3 shows the vertical nano-column grown by pulsed MOCVD method. During the pulsed growth mode the group III and group V precursors were introduced alternately in the sequence: TMGa, 1 sccm, 20 s; NH_3, 1 slm, 60 s. The grown GaN nano-columns were observed to be hexagonal symmetry of the sidewall facets as seen in Fig 3. High resolution FE-SEM image clearly show the perfect hexagonal symmetry. The results suggest that a highly oriented GaN nano-column could be grown by pulse MOCVD method. Further, Raman, photoluminescence (PL) and cathodoluminescence (CL) experiments are under investigation on this sample for more understanding which will be included during the time of conference.

3. Conclusions

In summary, a novel technique was used to grow GaN nano-columns on Au-coated Si by pulse flow MOCVD method. The Au coated Si substrate was annealed prior to GaN growth to obtain nano-columns in a vertical direction. From the SEM image, GaN nano-columns grown are vertical in direction and hexagonal in shape. The uniform shape and smoothness of the nano-column clearly indicates that it could be useful for device application.

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