ARTICLE IN PRESS

Superlattices and Microstructures xxx (2016) 1–6

Contents lists available at ScienceDirect

Superlattices and Microstructures

journal homepage: www.elsevier.com/locate/superlattices

Material characteristics of self-assembled mushroom-like InGaN nanocolumns

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ARTICLE INFO

Article history:
Received 16 December 2015
Received in revised form 8 March 2016
Accepted 12 March 2016
Available online xxx

Keywords:
InGaN
Mushroom-like
Nanocolumns
Molecular beam epitaxy

ABSTRACT

The material characteristics of self-assembled mushroom-like N-polar InGaN/GaN nanowire heterostructure have been clarified, which were achieved by different In content self-assembled InGaN nanocolumns grown on self-assembled GaN nanocolumns template on (111)-silicon-substrate under N-rich condition by plasma-assist molecular beam epitaxy (PA-MBE). The In component of the InGaN nanocolumns was determined by XRD (2θ-ω scans). SEM has been used to study the morphology which demonstrated that the diameter of the nanocolumns became larger with higher In content. The structural properties of the individual InGaN nanocolumn were further analyzed by HAADF image, EDX and TEM. The high-In-content (85%) single mushroom-like InGaN nanocolumn showed some cracks on the sidewall, however the GaN nanocolumns showed dislocation free. The (0002) facet of the nanocolumn show very clearly hexagonal structure. It is quite clear that the formation of the mushroom-like InGaN nanocolumns comes from that the lateral epitaxy is dominating with the high In content embedded.

1. Introduction

III-Nitride semiconductor-based structures have made a great success in optoelectronic device applications in the last 2 decades, due largely to the wide direct bandgap exhibited by GaN, which was emitting blue light and achieved the white light LED. At present, the indium gallium nitride ($In_xGa_{1-x}N$) compound semiconductor is considered as the most promising material system because of their superior material properties and the fact that the direct band gaps of $In_xGa_{1-x}N$ is tunable from the near-infrared (0.64 eV, InN) [1–5]to near-UV (3.4 eV, GaN). These reports suggest that high-In-content InGaN has the potential to realize high performance optoelectronic devices at the optical communication wavelength. Very recently, several groups have demonstrated the growth of InN-based heterostructures, aiming at device application [6–8]. However, the material quality of InN and high-In-content InGaN epitaxial layers was not sufficient for the heterostructure device because of the high density of threading dislocations and residual electrons. A proposed method to overcome the issue is to use semiconductor nanocrystal, since self-organized nitride nanocolumns [9,10] are almost dislocation free. This technology may bring about an improvement for the crystalline quality of InN and high-In-content InGaN. Furthermore, the nanocolumns exhibit many important advantages including low dislocation density, high electron mobility, and large saturation velocity

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http://dx.doi.org/10.1016/j.spmi.2016.03.018

Please cite this article in press as: Q.M. Chen et al., Material characteristics of self-assembled mushroom-like InGaN nanocolumns, Superlattices and Microstructures (2016), http://dx.doi.org/10.1016/j.spmi.2016.03.018

[11]. Anisotropic epitaxial growth of free-standing GaN [12,13] and InGaN NWs [14,15] presents a key technological advancement in material structure for light-emitter technology. The catalyst-free [16–18] and spontaneous growth of (N-polar) GaN nanowires [19] based on plasma-assistant molecular beam epitaxy (PA-MBE) eliminated the use of foreign metal, such as gold catalyst, which act as non-radiative recombination center and constitute material incompatibility with silicon foundry technology.

In this work, we report the growth of high-In-content InGaN nanocolumns on Si (111) substrates by PA-MBE. The tendency of becoming mushroom-like with the In content increasing was demonstrated by X-ray diffraction (XRD) and scanning electron microscopy (SEM). We also demonstrated the hexagonal-shaped top structure of single InGaN nanocolumn by TEM.

2. Experimental procedure

N-polar self-assembled GaN nanocolumns have been grown on Si (111) substrate under N-rich condition by PA-MBE. Special Knudsen cells were used for In and Ga metal sources to obtain good beam stability and a conventional rf-plasma cell was used to excite the nitrogen gases active. The n-type Si (111) substrate with a scale of 2 inch substrate was deoxidized for 40 s in (10%) HF. Then, it was mounted In-free on a sample holder and degassed at high temperature until the appearance on the reflection high-energy electron diffraction(RHEED) pattern of a clear 7 × 7 reconstruction, characteristic of a clean (111) Si surface. The chamber temperature was first raised to 900 °C for an *in situ* oxide desorption, which was keeping 30mins. The initial nitridation of the substrate surface at approximately 780 °C in the growth chamber, GaN dots were grown at approximately 780 °C as nucleation seeds of the nanocolumns. The V/III ratio was maintained 15 and growth rate up to 100 nm h^{-1} was observed. When the GaN nanocolumns were grown for 1 h, the InGaN nanocolumns were grown at approximately 550 °C for 2 h under a N-rich condition. We grew four InGaN samples a, b, c, d with the In component set at 30%, 35%, 44% and 85%, respectively. Fig. 1 shows a schematic diagram of InGaN/GaN nanowire structure. The height of four InGaN samples was about 200 nm. In/Ga beam flux and the nitrogen-plasma condition for depositing the GaN and InGaN nanocolumns were precisely controlled by a shutter control method so that the effective V/III ratio was as unity as possible in terms of stoichiometry, which was already confirmed to be essential to obtain good crystalline quality.

The structural properties of the self-assembled InGaN/GaN nanocolumns heterostructure were studied by XRD, SEM, EDX and TEM. For the EDX and TEM studies, the samples were prepared by transferring nanowires from the Si substrates to a Cu grid with carbon supporting film. The EDX data was collected with an Oxford Instruments plc detector and analyzed with INCA software.

3. Results and discussion

The In content of these four InGaN/GaN nanowire heterostructures were confirmed by XRD (0002) 2θ-ω measurement which was shown in Fig. 2(a). From the curve of the four samples, they all have the peak that 2θ equal to 28.1 coming from n-type Silicon substrate, which was a calibration of the In content. The peak corresponding to x-axis equal to 34.5 coming from the GaN nanocolumns is very sharp, which indicated that the GaN nanocolumns were grown well. Between the Si peak and GaN peak we can see the different InGaN peaks of the four samples. From the figure we can see the In content of InGaN nanocolumns was increasing. The In content increasing tendency of four samples accompany with the 2θ was shown in Fig. 2(b), which indicated the In content of the four samples a, b, c, d was 30%, 35%, 44%, 85%, respectively. In addition, from the sample d, there was a peak at 31.5 which indicated that the In content was 85%. This is evidence that the high In content InGaN self-assembled nanowires have been realized successfully. On the right of this peak, there was another low In content peak due to the phase separation of the high-In -content InGaN nanocolumns.

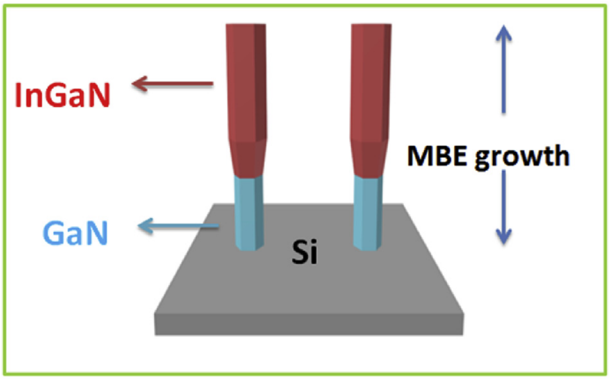

Fig. 1. Schematic diagram of the self-assembled InGaN/GaN nanocolumns on silicon substrate.

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