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Two step growth of high quality long n-GaN:Si nanowires using μ-GaN seed on Si(111) by metalorganic chemical vapor deposition

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ABSTRACT

A two-step growth method for growing high quality long n-GaN:Si nanowires (NWs) on Si(111) substrates using metalorganic chemical vapor deposition (MOCVD) was developed. In the primary step μ-GaN seeds were grown at 710 °C by pulsed growth method using MOCVD and in the secondary stage, we suitably increased the growth temperature to 950 °C in order to grow the high quality long n-GaN:Si NWs by continuous flow mode. We grew n-GaN:Si NWs at various pairs of μ-GaN seed so as to examine its effect on the growth rate. The density and length of n-GaN:Si NWs were improved with the increase of seeds up to 10 pairs. The number of seed pairs determines the density and length of n-GaN:Si NWs, but they did not affect its diameter directly. Field emission scanning electron microscopy, X-ray diffraction, photoluminescence, cathodoluminescence and high-resolution transmission electron microscopy were used to characterize the specimens.

1. Introduction

Semiconductor nanostructured materials have attracted extensive interest over the past decade due to their importance in fundamental research and the development of next generation devices [1–3]. As the potential building blocks for nano-electronic, nano-optical, and nano-mechanical devices [1], nanowires have received considerable attention from the scientific and engineering communities. Gallium nitride (GaN) with a direct and wide band bandgap of 3.39 eV at room temperature has been widely used in opto-electronic and high power high-frequency electronic devices. GaN is also a promising material for field emitters because of its low electron affinity as well as its excellent physical and chemical stabilities. Among gallium nitride materials, GaN nanowires (NWs) have attracted much attention in the past few years due to their potential applications in photonic and biological nanoscale devices such as blue light emitting diodes, short wavelength UV nano-lasers, and biochemical sensors [2–6]. The nanowire geometry improves the crystalline quality by decreasing the dislocation density and internal stress while enhancing the light guiding and extraction. The wires also serve as very interesting templates for obtaining uniaxial/coaxial InGaN/AlGaN-based heterostructures using free surface relaxation and different facet orientations [7–9].

To date, several methods such as, carbon-nanotube-confined reactions, laser ablation, molecular beam epitaxy and catalytic chemical vapor deposition have been reported to grow GaN NWs [10–12]. However, GaN NWs by metalorganic chemical vapor deposition (MOCVD) with high quality is seldom reported. In our previous work, we have grown GaN NWs by two different kinds of method using MOCVD. One is the growth of undoped GaN (μ-GaN) NWs by pulsed method at low temperature (740 °C ± 20 °C) having high density (11×10^7 cm^{-2}) and short length of 1.5 to 2 μm [13]. The other one is the non-pulsed growth method (continuous flow mode) at high temperature (900 °C) having low density and long length of 3 to 5 μm [14–16]. To overcome these limitations of low density and short length, we have combined these two kinds of growth method. In our present work, the n-GaN:Si NWs were grown by recently developed two-step growth method. The advantage of two-step growth method is that n-GaN:Si NWs were fabricated with improved density and length.

Here, the pulsed μ-GaN NWs grown at low temperature were called as pulsed GaN seed. Because, pulsed μ-GaN NWs at low temperature were used as seed for high quality n-GaN NWs at high growth temperature. Comparison with the density and length of the n-GaN:Si NWs grown on various pairs of μ-GaN seed were reported here. The grown n-GaN:Si NWs were characterized by field-emission scanning electron microscopy (FE-SEM), X-ray diffraction (XRD), photoluminescence (PL) and cathodoluminescence (CL) spectroscopy. The n-GaN:Si NW morphology was further analyzed by high-resolution transmission electron microscopy (HRTEM).

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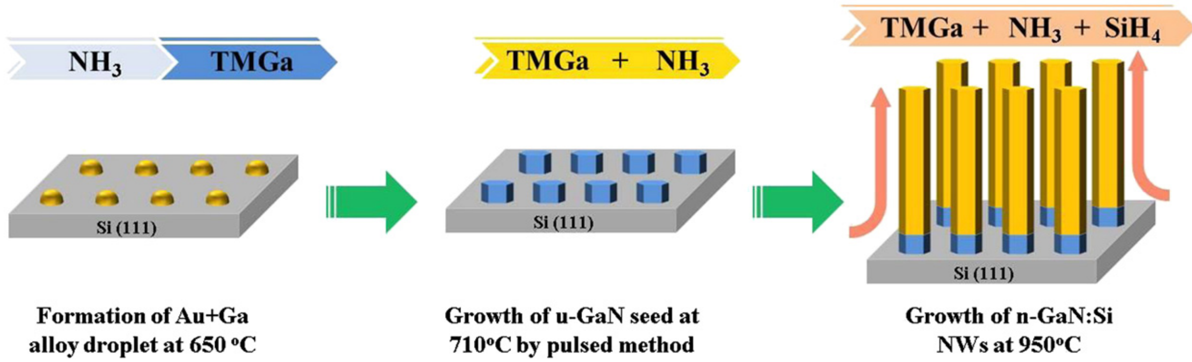

Fig. 1. Schematic diagram of n-GaN:Si NWs grown by 2-step method.

2. Experimental details

We have developed original growth method to get n-GaN:Si NWs on Si(111) substrate using MOCVD by a two-step growth process. Trimethylgallium (TMG), ammonia (NH_3), and silane (SiH_4, 10 ppm diluted in H_2) are the precursors for Ga, N and silicon respectively. Fig. 1 shows the schematic representation of experimental procedure for n-GaN:Si NWs on pulsed GaN seeds. The primary stage of the two-step growth process typically begins with a coated Au film on Si(111) substrate by ion sputtering system. The Au + Ga nano-droplets were formed by MOCVD under hydrogen ambient for 10 min at 650 °C with a reactor pressure of 600 Pa. This nano-droplets act as a nucleation seed for the growth of GaN, in which the utilized Au catalyst no longer exists at the top of NWs. After that, the pulsed GaN seeds were grown on Au + Ga nano-droplets using pulsed flow MOCVD. We prepared 4 kinds of different samples in order to analyze the effect of seeding. Pulsed μ-GaN seeds were grown up to 15 pairs at a working pressure and temperature of about 600 Pa, and 710 °C respectively. During the pulsed growth mode, the group-III and group-V precursors were introduced alternately in the following sequence: TMG, 3 min; NH_3, 3 min. In the last step, n-GaN:Si NWs were grown on pulsed μ-GaN seeds at 950 °C for 1 h with silane doping by non-pulsed mode. During the non-pulsed growth mode, the group-III and group-V precursors were introduced simultaneously. The seeding layer of pulsed GaN was undoped crystal, but the n-GaN NWs were grown with the doping of silane gas, which is a critical factor to initiate the vertical nanowire growth [17].

The morphology of the n-GaN:Si NWs was investigated by field emission scanning electron microscopy (FE-SEM, S-4800/HITACHI) between the operating voltages of 10.0–15.0 kV. Crystalline phases in the as-grown products were characterized by powder X-ray diffraction (XRD/X'pert-MRD/Philips) with monochromated Cu Kα radiation using 40 mA current and 45 kV respectively. In our present study, the Bragg–Brentano configurations were used for XRD measurements. The optical properties of the as-grown NWs were characterized by photoluminescence (PL) analysis using a 325 nm He–Cd laser and a cathodoluminescence (CL) attached to the FE-SEM system. Microstructure and growth directions were evaluated by the high-resolution transmission electron microscopy (HRTEM/JEM-2010/JEOL) at an operating voltage of 200 kV. The selected area electron diffraction (SAED) pattern and the lattice image were obtained by Inverse Fast Fourier Transform (IFFT) and Fast Fourier Transform image (FFT).

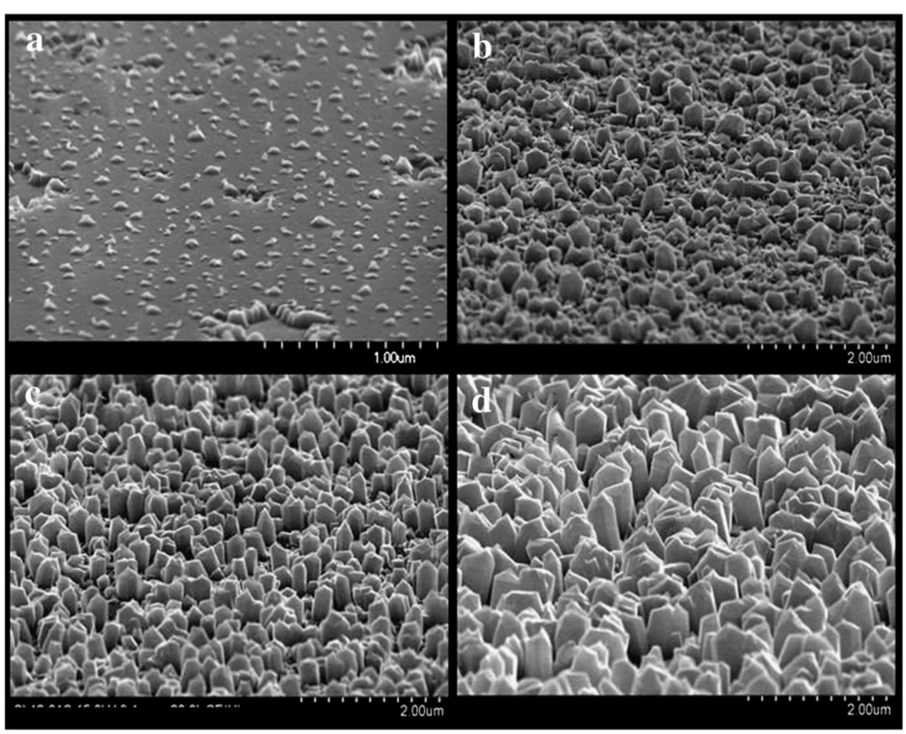

Fig. 2. Tilt-view FE-SEM images of pulsed GaN seed with 0 pairs (a) 5 pairs (b), 10 pairs (c), 15 pairs (d).