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Growths of indium gallium nitride nanowires by plasma-assisted chemical vapor deposition

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ABSTRACT

InGaN nanowires (NWs) were grown on Si(100) at 700 °C using Au catalyst in a plasma-assisted chemical vapor deposition reactor. As the indium vapor pressure was low around 16 mPa during the growths, only the curved GaN NWs could be grown containing indium impurities. By increasing the indium vapor pressure to 53 mPa during the growths, InGaN NWs were transformed to less curved NWs with a broad distribution of NW diameters from 20 to 90 nm. The room temperature photoluminescence of InGaN NWs grown at a high indium vapor pressure showed a broad emission peak at 417 nm, corresponding to an average of 14.5% indium composition in the NWs, with a large full-width at half maximum of 77 nm. Transmission electron microscopy characterization of InGaN NWs showed that the growth orientation was along [100] for the low indium vapor pressure growths and was transformed to along [001] for the high indium vapor pressure growths.

1. Introduction

The nitride semiconductor materials have great potentials in many applications such as light emitting diodes and lasers owing to their direct and wide band gaps. Gallium nitride has the band gap of 3.43 eV at the room temperature and relatively large exciton binding energy of 28 meV, making its special among nitride semiconductor materials. Compared with other devices, indium gallium nitride (InGaN) devices were characteristic of the wide wavelength range (1.12–3.43 eV) of emission in the visible light range [1]. To grow InGaN with a high indium content, the effects of growth temperature, hydrogen incorporation, and indium flow rate on the composition of InGaN nanowires (NWs) were studied [2,3]. The increase of indium content in the InGaN grown at a high temperature causes the formation of a large amount of dislocations and serious phase separation [4,5].

High-quality InGaN nanostructures are generally grown in plasma-assisted molecular beam epitaxy [1,6,7] and hydride vapor phase epitaxy [8]. The most common catalyst-assisted vapor–liquid–solid (VLS) and vapor–solid (VS) growth methods are still widely exploited for the synthesis of one-dimensional NWs. The advantage of VLS mechanism is the selective growth of NWs with a uniform diameter and has avoided the problems of lattice mismatch [9]. Recently, Yang et al. grow perfect InGaN NWs using the VS growth mechanism to emit a wide range of visible light.

The difficulty to grow the NWs with high indium contents and the poor quality of InGaN NWs grown at low temperatures due to insufficient ammonia dissociation remained as serious issues. In order to grow the InGaN NWs with high indium contents, the problem with the low degree of dissociation of ammonia at a low temperature must be solved. Therefore, the plasma-assisted chemical vapor deposition (PACVD) system, utilizing the plasma to dissociate nitrogen or ammonia to efficiently provide a high concentration of nitrogen radicals, is employed in this study. The PACVD system has the ability to produce high-quality catalyst-assisted InGaN NWs in the low temperature growths [10].

2. Experimental

InGaN NWs were grown on Si(100) substrate deposited with a film of gold catalyst in a PACVD system. The experimental equipment is schematically shown in Fig. 1. InGaN NWs were grown in a 33-mm quartz-tube inserted in a three-temperature-zone furnace. For the pretreatment of Si(100) substrates, the organic contamination on Si(100) surface was removed by the use of ethanol and acetone in an ultrasonic bath (30 min). Afterwards, the native oxide layer of Si was etched away by immersed HF solution for 1 min. Finally, the Si substrate coated with Au catalyst was placed at zone 1 center of the furnace and immersed in the nitrogen plasma during the reaction. The metallic gallium and indium droplets of 99.9999% purities used as the vapor sources of reaction were placed in the 15-mm quartz tube at zone 2 and zone 3 center of the furnace, respectively. In order to tune the indium composition in InGaN NWs, the indium droplet was operated at the various

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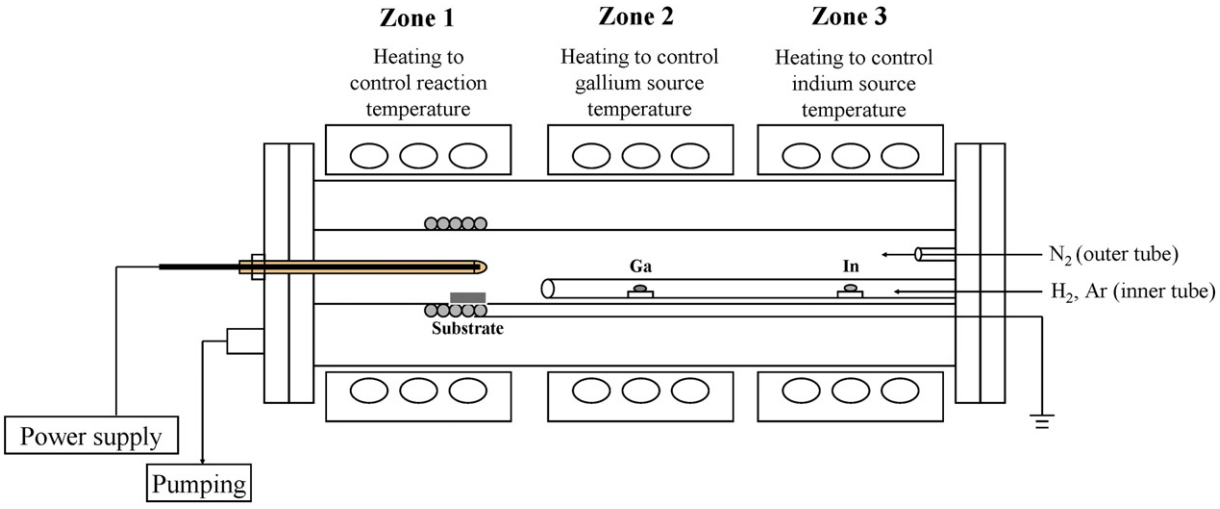

Fig. 1. A schematic diagram of the (In)GaN nanowires growth apparatus. The substrate (zone 1), immersed in the N_2 plasma, metal gallium (zone 2) and indium (zone 3) droplets were placed in 33 mm and 15 mm quartz tube, respectively.

temperature of 750 °C, 800 °C and 850 °C as the substrate and gallium droplet were respectively controlled at 700 °C and 900 °C. H_2 (40 sccm) and Ar (250 sccm) were mixed with the N_2 carrier gas (160 sccm) to carry the metal vapors into zone 1 at the working pressure of 266 Pa. To avoid the formation of GaN or InN layer on the surface of metal droplets during their exposure to nitrogen plasma, nitrogen was injected into the 33-mm quartz tube (outer tube) rather than the 15-mm quartz tube containing metal droplets. In order to averting the violent gas phase reaction of metal atoms and nitrogen free radicals, H_2 was used as the reducing gas. The use of Ar facilitated the plasma generation and was able to enhance the plasma density and the decomposition of nitrogen gas. A 25-kHz high-frequency power supply (Creating Nanotechnology Inc.) was used to generate nitrogen plasma at zone 1 utilizing the principle of dielectric barrier discharge.

In order to tune the indium composition of the InGaN NWs, the increase of indium vapor pressure was from 16 to 200 mPa by dominating the indium droplet temperature from 750 °C to 850 °C. The surface morphology of the InGaN NWs was analyzed by the field emission scanning electron microscopy (FESEM, Philips XL-40FEG) at a voltage of 10 kV. The structure and crystal orientation of NWs were further analyzed by the field-emission transmission electron microscopy operated at a voltage of 200 kV (HR-TEM, JEOL JEM-2100 F) as well as by X-ray Diffractometer (XRD, D8 DISCOVER, Bruker AXS Gmbh, Cu-Kα) with a fixed incident angle of 1° under the operated voltage of 40 kV. The photoluminescence (PL, ULVAC/Jobin Yvon/Labram HR) spectroscopy with He-Cd laser source (20 mW) of 325 nm was utilized to characterize the band-to-band emission of InGaN NWs at room temperature.

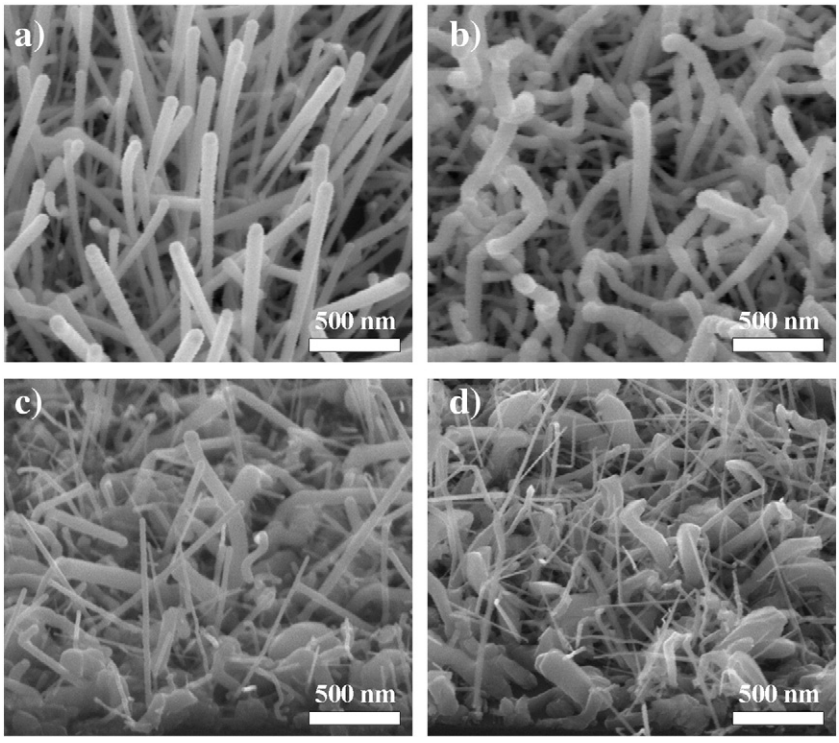

Fig. 2. SEM image inclined angle of 45° of the as-grown (In)GaN nanowires on the Au-coated Si(100) grown at 700 °C and 266 Pa. The (In)GaN nanowires were grown at the various indium vapor pressure of (a) 0 mPa (Pure GaN NWs) (b) 16 mPa (c) 53 mPa (d) 200 mPa.