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Synthesis of three kinds of GaN nanowires through Ga₂O₃ films' reaction with ammonia

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Abstract

A new method was employed to obtain GaN nanowires (NWs). In this method, SiC films were deposited with radio frequency (r.f.) magnetron sputtering onto silicon substrates and annealed at high temperature, and then Ga_2O_3 films were deposited on top of the SiC intermediate layers and annealed in NH₃ atmosphere. SiC layer was used to reduce thermal and lattice mismatch between GaN and Si, and improve NWs' quality. After Ga_2O_3 films reacted with NH₃, a great quantity of GaN NWs with the shape of birch trunks and stalactites were found by transmission electron microscopy (TEM). At the same time, a few very even and uniform pillarlike NWs were observed. The electron diffraction patterns (EDP) show that birch trunk-shaped and pillarlike NWs are all single-crystalline structures. These NWs were also analyzed with the assistance of X-ray diffraction (XRD), Fourier transformed infrared spectra (FTIR) and high-resolution transmission electron microscopy (HRTEM) to show their properties.

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Keywords: GaN; Intermediate layer; Stalactite-shaped; Birch trunk-shaped; Pillarlike

1. Introduction

As a kind of important semiconductor, GaN attracted extensive attention for decades. GaN

nanowires (NWs), nanorods and nanobelts have promising device applications in 1-D systems due to their excellent properties. Many groups have prepared GaN on the nanoscale. Chia-Chun Chen et al. produced high-quality GaN NWs using vapor–liquid–solid (VLS) technique [1]. Joushua Goldberger et al. formed single-crystal GaN nanotubes with the assistance of template [2].

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H. Li et al. fabricated bamboo-shaped GaN nanorods in 2002 [3]. Other methods such as thermal chemical vapor deposition (CVD) [4], oxide-assisted technique [5], sublimation [6], arc discharge [7], laser ablation [8], and pyrolysis [9] were also utilized to produce GaN NWs. Currently, Höche et al. reported the preparation of well-ordered substrate-adhered GaN NWs with diffraction-mask-projection excimer-laser patterning [10]. A novel method was carried out to prepare single-crystalline GaN NWs in our laboratory [11]. Only several simple appliances were used in this method, so it simplified GaN NWs' preparation technique greatly. However, the NWs we got are not even and smooth, and a big thermal mismatch between GaN and silicon substrate will restrict GaN NWs' application in devices. In order to avoid this defect and improve GaN's quality, we used SiC as the intermediate layer.

2. Experimental details

In our experiment, we formed GaN NWs by self-assembling of Ga_2O_3 films in their reaction with NH₃. The films were deposited by magnetron sputtering on the SiC intermediate layer on silicon substrates.

The first step is the process of forming SiC intermediate layers. SiC films were deposited to silicon substrates with a JCK-500A radio frequency (r.f.) magnetron sputtering machine. Clean Si (111) wafers were employed as substrates. SiC wafers with the purity of more than 99.999% were used as targets. The conditions of sputtering were as follows: the background pressure was 4×10^{-4} Pa; the distance between targets and substrates was 8 cm; the pressure of Ar (\geq 99.99%) was 2 Pa; the sputtering power was 150 W and the frequency was 13.56 MHz. After 15 min, SiC films with a thickness of about 50 nm were obtained.

Subsequently, SiC films were put into the anneal system (Fig. 1). The furnace was kept at 950 °C. N₂ (\geq 99.999%, with a flow of 1000 ml/min) was introduced into the oven chamber to protect the samples. SiC intermediate layers with the same thickness formed at high temperature after 15 min.



Fig. 1. The anneal system, also the reaction system.

The second step is to deposit Ga₂O₃ films and synthesize GaN NWs. The SiC-covered wafers were sputtered with Ga₂O₃ (99.999%) for 90 min under the same conditions and 500 nm thick films consisting of Ga₂O₃ nanoparticles were obtained. Then the samples were placed into the reaction system without the least delay. N2 was introduced into the system for 5 min to expel the air. Subsequently, NH₃ (99.999%) with a flux of 500 ml/min was introduced into the system. The reaction lasted for 5, 10 and 15 min, respectively. And the corresponding samples were named A, B and C. As our observation, Ga₂O₃ began to convert into GaN at about 800 °C. The reaction equation expressed can be as $Ga_2O_3 +$ $2NH_3 = 2GaN + 3H_2O\uparrow$.

After these processes, three kinds of GaN NWs were obtained. We analyzed the samples with Rigaku D/max-rB X-ray diffraction (XRD) meter (Tokyo, Cu K α , $\lambda = 1.54178$ Å, 2θ mode, $30^{\circ}-50^{\circ}$) at room temperature to specify their crystalline structure. Nicolet710 Fourier transformed infrared spectra (FTIR) meter was used to measure the samples' chemical states. Transmission electron microscopy (TEM) (Hitachi H-800) and high-resolution transmission electron microscopy (HRTEM) (GAM2010, JEOL company) were carried out at room temperature to measure the samples' morphology and microstructure, respectively.

3. Results and discussion

3.1. XRD analysis

The XRD pattern of sample C is depicted in Fig. 2. Peaks were found at $2\theta = 32.3^{\circ}$, 34.5° and

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