Journal of Alloys and Compounds 723 (2017) 21-29

Contents lists available at ScienceDirect

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Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Temperature effects for GaN films grown on 4H-SiC substrate with 4° miscutting orientation by plasma-assisted molecular beam epitaxy



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ARTICLE INFO

Article history: Received 11 May 2017 Received in revised form 20 June 2017 Accepted 21 June 2017 Available online 24 June 2017

Keywords: Molecular beam epitaxy Gallium nitride Silicon carbide Post-annealing Compound semiconductor

ABSTRACT

The effects of growth temperatures and post-annealing treatment for GaN films by plasma-assisted molecular beam epitaxy are investigated. The heteroepitaxial GaN films were deposited on 4H-SiC substrates with 4° miscutting orientation at growth temperatures from 700 °C to 800 °C with a constant N/Ga flux ratio, while the post-annealing process was carried out at 800 °C for 10 min. GaN films are characterized by reflective high-energy electron diffraction, field-emission scanning electron microscopy, atomic force microscopy, X-ray photoelectron spectroscopy, high-resolution X-ray diffraction and photoluminescence spectroscopy. For the growth at the temperature of 750 °C, we can obtain a smooth surface, high percentages of Ga-N bond and low $R_{GaO/GaN}$ revealing a more stable composition on the surface. The higher crystalline structure of GaN films can be also obtained with the minimum of threading dislocation density. The sharpest near band edge emission and lowest defect band emission can be observed for GaN films grown at temperature of 800 °C. However, the surface roughness increases, and surface decomposition of GaN films occurs for the high-temperature growth. Moreover, post-annealing process can make the surface smoother, remove Ga droplets, and improve the stability of surface composition. After the post-annealing process, the increase of crystalline quality and optical property can be also demonstrated by the reduction of dislocation density and yellow band emission, respectively. In summary, the optimization of growth temperatures and post-annealing process can produce a high-quality GaN compound semiconductor for the applications in the future.

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1. Introduction

Gallium nitride (GaN) compound semiconductor is one of the most promising materials, which has excellent properties such as wide band-gap, high electron mobility, good thermal conductivity and high temperature stability [1,2]. Therefore, GaN films has been applied in optoelectronic and electronic devices such as light emitting diodes (LEDs) [3–5], solar cells [6–8], sensors [9,10] and transistors [11]. However, high quality and low growth temperature requirement of GaN films is an important and challenging issue in the fabrication of GaN-based devices [12,13].

Various epitaxial techniques have been devoted to control the high quality of GaN films. Plasma-assisted molecular beam epitaxy (PAMBE) is a simple and low-temperature technique for the growth of GaN films by controlling the growth parameters [14]. Several

* Corresponding author. E-mail address: isyu@mail.ndhu.edu.tw (I.-S. Yu). relative studies have been done to investigate GaN nanostructures grown on sapphire substrate via the growth temperatures by PAMBE [15,16]. Besides, the post-annealing process is also an interesting topic to optimize GaN films for their applications. Li et al. reported that thermal annealing can improve the crystal quality of GaN epilayers grown by MBE [17], while Cheng et al. investigated that post-annealing treatment of GaN films by pulsed laser deposition (PLD) has improved not only the films surface and crystalline structure, but also obtained strain-free GaN films [18]. However, the original problem, lattice mismatch and incompatibility of thermal expansion coefficient between GaN films and substrates, is a serious concern for determining the growth mechanism of GaN epitaxial layers. SiC substrate is a promising candidate for GaN epitaxial layers due to smaller lattice mismatch (3.1%) and higher thermal conductivity for the heat dissipation of GaN-based devices [19]. Further, the miscutting orientation of SiC substrates also has advantages for the growth of III-nitride based semiconductors. Tanaka et al. reported that the misorientation of SiC from (0001) plane can enhance two dimensional growth of

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Table	1

Su	mmarv	of GaN fil	lms grown o	n 4H-SiC substrat	es at the differen	nt growth tempe	rature and in-situ	post-annealing pro	ocess

Sample	Growth temperature	Growth time	Ga beam equivalent pressure	Ga K-cell temperature	Post-anneal temperature	Anneal time
T1	700 °C	1 h	3.5×10^{-7} Torr	900 °C	x	x
T2	750 °C	1 h	$3.5 imes 10^{-7}$ Torr	900 °C	х	х
T3	800 °C	1 h	3.5×10^{-7} Torr	900 °C	х	х
T4	700 °C	1 h	3.5×10^{-7} Torr	900 °C	800 °C	10 min

epitaxial AlN by reducing the distance which atoms must diffuse on the substrate surface to incorporate into the crystal [20]. Smith et al. presented that the misorientation at the angle of 4° can relieve the stress at the interface between 6H-SiC substrate and GaN films [21]. So far, a detailed study for various growth temperatures on 4H-SiC (0001) with 4° miscutting orientation by PAMBE has not been reported yet. The correlation of temperature effects and GaN films grown on this special substrate is needed to be investigated.

In this report, we study GaN films grown in the parameters of different substrate temperatures and post-annealing treatment on 4H-SiC substrates with 4° miscutting orientation by PAMBE. The surface condition and chemical compositions of GaN films will be investigated by reflective high-energy electron diffraction (RHEED), field-emission scanning electron microscopy (FE-SEM), atomic force microscopy (AFM) and X-ray photoelectron spectroscopy (XPS), whereas the crystalline structure and optical properties of GaN films will be studied by using high-resolution X-ray diffraction (HRXRD) and photo-luminescence (PL) spectroscopy, respectively. We have demonstrated the study for more understanding about the

influences of substrate temperatures and post-annealing process on the properties of GaN films grown on 4H-SiC substrates with 4° mis-cutting orientation.

2. Experimental

The growth of GaN films on 4H-SiC (0001) substrate was carried out by ULVAC PAMBE system equipped an *in-situ* RHEED. The detail of substrate preparation, growth configuration, and processing parameters for the samples was similar to the previous report [22]. According to the report, GaN films were grown at a fixed N/Ga flux ratio of 28 at three different substrate temperatures: 700, 750 and 800 °C, respectively. In order to investigate the effect of *in-situ* thermal annealing treatment, the post-annealing process was carried out at the temperature of 800 °C and the pressure of 4.5×10^{-8} Torr for 10 min after growth of GaN films. During the MBE growth, the beam equivalent pressure (BEP) of nitrogen plasma was fixed at 9.7×10^{-5} Torr, and Ga BEP of 3.5×10^{-5} Torr was provided by the K-cells at the temperature of 900 °C. These



Fig. 1. RHEED patterns: (a) 4H-SiC (0001) substrate, (b) GaN film grown at 700 °C, (c) GaN film grown at 750 °C, (d) GaN film grown at 800 °C, and (e) GaN film grown at 700 °C and post-annealing process at 800 °C.

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