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Effects of substrate and annealing on GaN films grown by plasma-assisted molecular beam epitaxy

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ABSTRACT

GaN films were deposited by plasma-assisted molecular beam epitaxy on different substrates, i.e. Si (111), Sapphire (0001) and 4H-SiC (0001) with 4° of miscutting orientation. Due to the lattice mismatch between GaN films and substrates, the substrates profoundly affect the growth mechanism of GaN films on them during the epitaxy. The smooth and complete GaN films can be obtained on 4H-SiC substrates based on the analyses of reflection high energy electron diffraction, scanning electron microscopy and atomic force microscopy. Higher quality of GaN films on 4H-SiC was also confirmed by high resolution X-ray diffraction. Photoluminescence measurements show that the compress stress existed in GaN films grown on sapphire, but tensile stress was observed in the films grown on 4H-SiC and Si. Meanwhile, *in-situ* annealing treatment of post expitxy can remove Ga droplets on GaN films and decrease the surface roughness and defect density of GaN films; however, thermal decomposition on the surface of GaN films was observed in the characterizations of X-ray photoelectron spectroscopy.

1. Introduction

Gallium nitride (GaN) semiconductor is one of the popular functional materials due to its extensive applications in optoelectronic and electronic devices. For the optoelectronic devices, GaN-based light-emitting diodes (LEDs) have been used in our daily life now, and their performance keeps improving by innovation of GaN growth and device design [1–3]. For example, high-performance GaN-based LED materials and devices have been tried to be realized on different substrates to fit their applications in the future [4]. For the electronic devices, due to its wide-bandgap property, AlGaN/GaN high electron mobility transistors (HEMT) have been applied in the high voltage and high temperature devices for microwave communications and RADAR [5–8]. For the applications in photovoltaics, heterojunction solar cells of GaN on silicon (Si) have some advantages, such as excellent blue response, simple processing steps and low processing cost [9–11]. GaN nanostructures have been pointed out to serve as photoelectrodes for photoelectrochemical water splitting to more efficiently generate hydrogen gas [12,13]. Moreover, GaN semiconductor has also been applied for sensor devices due to its piezoresistive and piezoelectric characteristics [14,15].

No matter what kinds of as-mentioned applications, it is required to have high quality of GaN films, so that different methods have been proposed to obtain high-quality GaN films. For example, high-quality GaN thin films can be deposited on sapphire with buffer layers for LED applications. In addition, many epitaxial techniques were proposed to grow high-quality GaN films, such as hydride vapor phase epitaxy (HVPE) [16], metal organic chemical vapor deposition (MOCVD) [17], molecular beam epitaxy (MBE) [18], pulsed laser deposition (PLD) [19], and the combination of MBE and PLD [20]. MOCVD has the high growth rate for industrial fabrications, but needs higher growth substrate temperature. Recently, PLD technique can make the epitaxial growth of GaN on thermally active substrates at low temperature [21]. The GaN growths by MBE technique also can be conducted at relatively lower temperature and obtained higher quality. For the growth of GaN nanostructures (nanorods, nanowires and nanodots), plasma-assisted MBE provides a simple means to fabricate self-assembled nanostructures without catalysts. The formation of GaN nanostructures can be controlled by the substrate temperatures, Ga/N ratio, growth time and substrates [22–26]. Besides the epitaxial methods, the choice of substrates is also an important issue to obtain high-quality GaN films for the application in various devices. For the epitaxy of GaN films, lattice mismatch and incompatibility of thermal expansion coefficient between epi-layer and substrate profoundly affect the growth mechanism and physical properties of GaN films [27–31]. As we know, annealing process is also an important key to produce blue and UV light emitters as well as high temperature devices. After the growth of GaN films, optimized post-annealing process can let us obtain highly crystalline and

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Table 1
Growth parameters of six samples.

Sample	Growth temperature	Growth time	Ga BEP (cell temperature)	Anneal temperature	Anneal time
Si	700 °C	1 h	4.6×10^{-5} Pa (900 °C)	X	X
Al_2O_3	700 °C	1 h	4.6×10^{-5} Pa (900 °C)	X	X
SiC	700 °C	1 h	4.6×10^{-5} Pa (900 °C)	X	X
Si-A	700 °C	1 h	4.6×10^{-5} Pa (900 °C)	800 °C	10 min
Al_2O_3-A	700 °C	1 h	4.6×10^{-5} Pa (900 °C)	800 °C	10 min
SiC-A	700 °C	1 h	4.6×10^{-5} Pa (900 °C)	800 °C	10 min

strain-free GaN films in order to improve the physical properties of GaN-based devices [32–34].

In this report, we investigated the substrate effects (i.e. Si (111), Sapphire (0001) and 4H-SiC) on the growth of GaN film by PAMBE as well as the film evolution under *in-situ* annealing treatment at 800 °C for 10 min in high vacuum condition. The growth parameters of substrate temperature, Ga/N ratio, and growth time were fixed at 700 °C, 0.035, and 60 min, respectively. To do so, we characterized the surface morphology, crystal orientation, defect density, residual strain, optical property, and surface composition of these samples. The surface condition of the substrates and epi-layers were investigated by reflection high energy electron diffraction (RHEED), atomic force microscopy (AFM) and scanning electron microscopy (SEM). High resolution X-ray diffraction (HRXRD) was also conducted to determine the crystal orientations and defect densities of the GaN films grown on these three substrates. Their residual stress and optical properties were studied by photoluminescence (PL) spectra. X-ray photoelectron spectroscopy (XPS) was used for the analysis of surface composition of the GaN epilayers.

2. Experimental

The growth of GaN films was carried out by an ULVAC PAMBE system. All of substrates were cleaned by acetone to remove organic impurity. In addition, Si (111) was cleaned by 10% HF solution to remove the native oxide and 4H-SiC was cleaned by H_2SO_4 and H_2O_2 (3:1) solution and 10% HF solution. After chemical cleaning, the substrates were loaded into the MBE chamber immediately. Until the base pressure of growth chamber lower than 1.0×10^{-7} Pa, thermal cleaning of Si, sapphire and 4H-SiC substrates were conducted at 850 °C, 850 °C, and 700 °C for 60 min, respectively. The growth parameters of all the samples are summarized in Table 1. The nitrogen plasma source was operated under the conditions of 500 W of power and 0.8 sccm of 6 N N_2 gas flow, which corresponds to 1.3×10^{-5} Pa of beam equivalent pressure (BEP). To study the effect of in-situ thermal annealing treatment for GaN films on different substrates, annealing process was at temperature of 800 °C and pressure of 6×10^{-6} Pa for 10 min after the growth of GaN films.

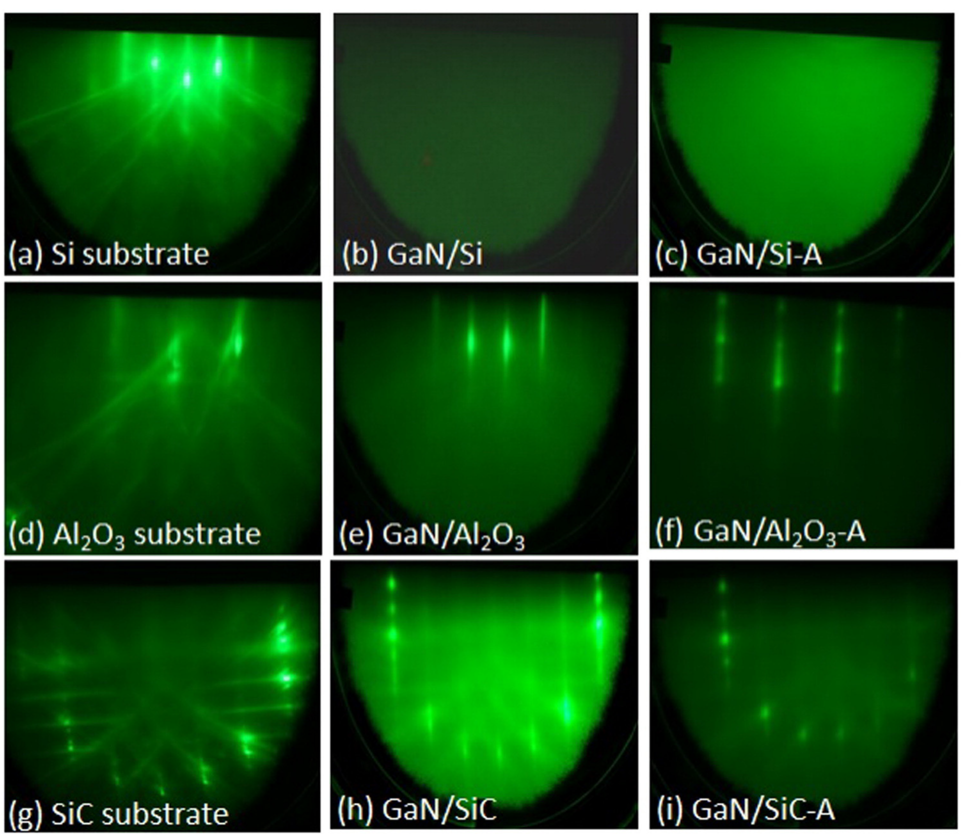

Fig. 1. RHEED patterns: (a) Si (111) substrate, (b) GaN film on Si as deposited, (c) GaN film on Si after annealing, (d) sapphire (0001) substrate, (e) GaN film on sapphire as deposited, (f) GaN film on sapphire after annealing, (g) SiC (0001) substrate with 4° miscutting orientation, (h) GaN film on SiC as deposited, and (i) GaN film on SiC after annealing.

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