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Influence of active nitrogen species on surface and optical properties of epitaxial GaN films

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ABSTRACT

Influence of active nitrogen species on surface and optical properties of homoepitaxial GaN films grown on GaN epilayers has been investigated. The epitaxial GaN films were grown at varying plasma powers (350–500 W) under identical growth conditions. High resolution X-Ray diffraction, Field Emission Scanning Electron Microscopy, Atomic Force Microscopy and Photoluminescence measurements were employed to characterize the structural, morphological and optical properties of the grown GaN films. High plasma power (500 W) lead to an increment in active nitrogen radicals and yielded high crystalline quality with reduced dislocations compared to low plasma power (350, 400 W) which divulge the presence of metallic gallium on the surface and low roughness. The valence band maximum position, electron affinity and ionization energy of the films increased with increment in plasma power. PL measurements revealed narrow and intense band to band edge emission with negligible defect related yellow band peak for the sample grown at 500 W. The analysis conveyed that higher amount of active nitrogen species encouraged good optical properties with insignificant defect states which can be employed for the fabrication of high performance optoelectronic & photovoltaic devices.

1. Introduction

III-Nitride semiconducting materials have gained huge attention in the field of optoelectronics and photonics [1–3] due to their noteworthy properties like wide direct band gap, high thermal conductivity and good thermal stability [4]. Plasma-Assisted Molecular Beam Epitaxy (PAMBE) technique has been employed for the growth of III-Nitride material system for subsequent design and development of efficient devices from past few decades. PAMBE growth offers very high crystalline quality, deposition at lower temperature, sharp interfaces and smooth surfaces with a precise control over the growth parameters [4–8]. Radio frequency (rf) inductively coupled plasma source has extensively been used as nitrogen activator source for the production of active nitrogen species [4,6]. The rf-plasma sources are advantageous as they create higher amount of favourable active nitrogen species (N or N*) compared to the molecular nitrogen (N_2 or N_2^+) [5]. It was reported that the rf-plasma sources enables high growth rate, low ion damage, high temperature operation and superior optical properties [9–11]. Jmerik et al. reported that the intensity of the flux of activated nitrogen originated from rf-plasma source can be linearly controlled, and an increment in the rf-plasma power lead to a higher amount of active nitrogen species with high growth rate [12]. Various studies have been performed to understand and analyse the role of nitrogen species on the growth of GaN films [5,12–14]. McSkimming et al. demonstrated that the growth rates of GaN films grown by PAMBE can be tailored and a growth rate of 2.6 μm/h can be achieved by varying the plasma conditions [15]. Tarsa et al. reported that the structure and morphology of the GaN films grown via PAMBE is extremely sensitive to the III/V ratio by varying the nitrogen flux [16]. The surface structure can be varied from Ga-stable regime (flat) to an intermediate or N-stable (island) growth regime. A strong dependence and direct correlation between deep level defect densities with plasma induced atomic and ionic nitrogen species was also reported [17].

In the present study, we report the influence of rf-plasma power generated active nitrogen species on the structural and optical properties of homoepitaxial GaN films. Surface properties were

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scrutinized via microscopic measurements (FESEM, AFM). Structural defects, surface morphology, and optical properties of the grown films were thoroughly analysed and correlated. The electronic structure and oxygen chemisorption is probed via X-Ray and Ultraviolet photoemission spectroscopy (XPS and UPS).

2. Experimental

Ultrahigh vacuum (UHV) based Riber compact 21 PAMBE system, operating at base pressure in the range of 2 E-11 Torr was used for the growth of epitaxial GaN films. An rf-plasma source (Addon) was attached to provide the active nitrogen species during the growth. Gallium (Ga) flux was supplied via heating the standard Knudsen cells to 980 °C, which resulted in a beam equivalent pressure (BEP) of 1.2 E-6 Torr. The homoepitaxial growth of GaN films were performed on commercially purchased MOCVD grown 3.5 μm thick GaN epilayers on c-plane sapphire substrate (MGcS). The ex-situ wet chemical cleaning of the substrates was performed via standard cleaning procedure [18]. The GaN films (thickness varied from 0.8 to 1.1 μm) were grown at a constant substrate temperature of 735 °C with plasma power varying from 350 W to 500 W, keeping all other parameters as constant. The samples are abbreviated as S1, S2 and S3 for films grown at 350, 400 and 500 W respectively.

Various characterization tools have been employed ex-situ to analyse the quality of the homoepitaxially grown GaN films. The crystalline quality has been ascertained by high−resolution x-ray diffraction (HRXRD, Pananalytical, X'Pert PRO MRD System) while the morphological and topographical properties were examined by using field-emission scanning electron microscopy (FESEM, ZEISS AURIGA) and atomic force microscopy (AFM, V-Veeco). XPS and UPS (Omicron Multiprobe Surface Analysis System) measurements were carried out to scrutinize the surface chemistry, energy band structure and Fermi level (FL) position of the grown films. Monochromatized AlKα (1486.7 eV) and He (I) (21.2 eV) radiation source was employed for XPS and UPS measurements, respectively. Using He-Cd laser as an excitation source of 325 nm, the room temperature (RT) photoluminescence (PL) has been performed on the samples to examine the optical properties and presence of defect states in the grown samples.

3. Results and discussion

Fig. 1 (a) shows the 2 theta−omega scan of the grown GaN film taken along (0002) plane representing the first and second order diffraction peaks of GaN and sapphire. The sharp peaks revealed the high crystalline quality of the grown epitaxial GaN film. Fig. 1 (b) represents a couple of curves plotted between the plasma power and the full width at half maximum (FWHM) of X-Ray rocking curves (RC) taken along (002) and (102) plane of diffraction given by (▲) and (★) symbol in the plot which will give a measure of screw and edge dislocations respectively. Since, the screw dislocations are developed by following a loop of atoms around dislocation line resulting into one plane up or down in the lattice and can be propagated from the template to the MBE grown GaN films. Thus, the variation in FWHM is almost negligible in RC taken along (002) plane from S1 to S3. However, the FWHM value of peak in RC taken along (102) plane was significantly reduced from Ga − stable to intermediate regime as the edge dislocations were formed due to growth of an extra half plane of atoms into the crystal lattice and cannot propagate into the overgrown film. Tsai et al. also reported that edge dislocations are dominant in Ga-stable growth regime [19]. The obtained screw and edge dislocation densities were further integrated to evaluate the total threading dislocation density (TDD) in the grown GaN films [18]. Hence, via using the FWHM values measured from RC of grown samples, the TDD was calculated to be 2.65×10^7 cm^{-2}, 2.62×10^7 cm^{-2} and 2.4×10^7 cm^{-2} for S1, S2 and S3 respectively. It was observed that the TDD for S3 was least when compared to other GaN films. Further, to evaluate the effect of reduced TDD at varied plasma power on the topographical properties of the grown film, the surface of the films has been probed by using FESEM and AFM.

Recent report suggested that the surface structure of the GaN films are highly affected by the Ga/N flux (i.e. plasma power at constant Ga flux) [14]. Fig. 2 (a−c) represent the surface morphology of the grown GaN films (S1 to S3) examined by FESEM displaying the step-flow growth. S1 shows large amount of small hexagonal pits (having an average pit size of ~50 nm) with metallic Ga droplets on its surface. The hexagonal pits are formed due to the existence of screw dislocations and indicate that the films grown on template are epitaxial wurtzite GaN (0001) films [20]. In the case of S2, the amount of metallic Ga droplets has been reduced which was attributed to the consumption of excess metallic Ga by the increased amount of active nitrogen species to form GaN with reduced pit density. However, the average size of the pit was observed to be doubled (~100 nm). On further increasing the plasma power to 500 W, there were no traces of metallic Ga droplets, though a few bigger hexagonal pits (shown in the right-bottom inset of Fig. 2 (c)) possessing an average size of ~200 nm were observed. The increase in average pit size infers that stress relaxation might occur during the transition from Ga - stable to intermediate regime with increasing plasma power. The surface topography of the GaN films were also scrutinized using AFM measurements. The 1 × 1 μm^2 AFM images (Inset of Fig. 2 (a−c))

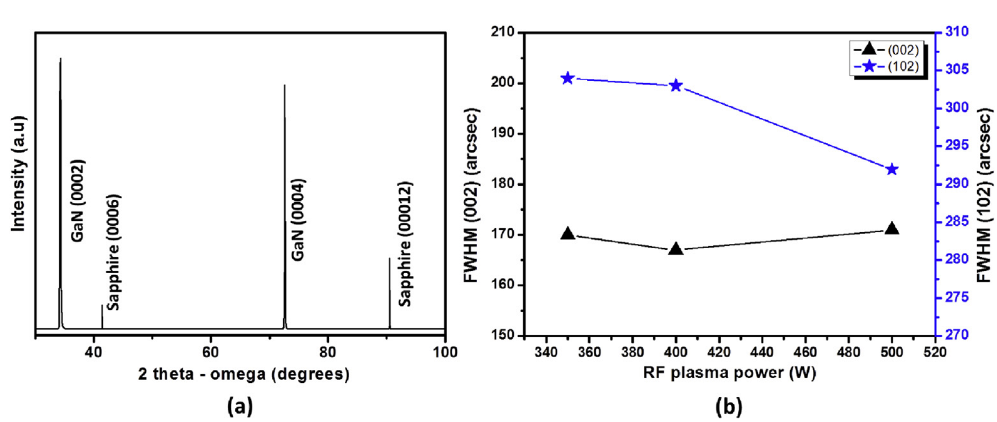

Fig. 1. (a) The 2-theta−omega scan of GaN film grown by rf-MBE at a plasma power of 500 W; (b) Plots of X-ray RC along (002) (▲) and (102) (★) plane w. r.t. rf plasma power.