

RF MAGNETRON SPUTTERING OF GALLIUM NITRIDE (GaN) ON SAPPHIRE SUBSTRATE

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The aim of this study is to determine the morphological properties and structure of GaN thin films deposited by RF magnetron sputtering grown on Al₂O₃ (0001). Moreover, it is crucial to find the most appropriate deposition conditions which lead to defect free GaN films. In pursuit of understanding the role of the epitaxial strain on the microstructural and morphological properties and the influence of the chemical composition of the target on GaN deposited films, different characterization methods are used. Atomic force microscopy, scanning electron microscopy are used for surface characterization and high-resolution X-ray diffraction is used for microstructural analysis of samples.

Keywords: Gallium nitride (GaN), thin film growth, RF sputtering

1. Introduction

The III-IV nitride semiconductors present nowadays a high interest for optical devices since the material defects do not have a big influence on their optical properties [1]. Particularly, the gallium nitride has shown impressive properties, because of its thermal stability and wide bandgap (3.4 eV) [2], for high temperature electronics, laser diodes, light emitting diodes, transistors, and high density optical storage. This semiconductor has two crystalline structures: hexagonal (wurtzite) and cubic (zinc-blende). Most of the research is done on the wurtzite phase form of GaN growth on the sapphire substrate, but the zinc-blende structure is also of interest because of its superior electronic properties. Due to the isotropic phase of the gallium nitride with cubic lattice, the phonon scattering is lower, therefore it is expected to have a higher saturated electron drift velocity [3].

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In RF magnetron sputtering, the crystallization process during which atoms undergo bonding rearrangements allow them to be transferred from a metastable phase to a crystalline phase using the substrate as a template. It is possible to sputter insulating materials, non-metals, oxides and dielectric substances [4]. This technique alternates the electrical potential of the current (reduces arcing and charge-up effects) by radio frequencies [5]. It is important to say that without using a magnetron, the non-conductive target materials become positively charged and the forming of thin film is more difficult. A main advantage of the RF-magnetron sputtering is that the plasma is capable of running in a pressure range 1-15 mTorr, therefore there is no need of secondary electrons to maintain the plasma.

Current reports of producing GaN films indicate that heteroepitaxial GaN films are grown on different substrates such as Si, Al₂O₃, ZnO, TiO₂, SiC, with different orientations [6,7]. To date, several synthesis methods were employed for the synthesis of GaN thin film by molecular beam epitaxy (MBE) [8,9], hydride vapor phase epitaxy (HVPE) [10], metal-organic chemical vapor deposition (MOCVD) [11,12], pulsed laser deposition (PLD) [13][14], low pressure chemical vapor deposition (LPCVD) [15], etc.

The stable phase of gallium nitride is the α -phase wurtzite structure. However, epitaxial layers can be achieved with the coexistence of wurtzite and zinc-blende (β -phase) phases due to the stacking sequence of nitrogen and gallium atoms. Both structure phases have polar axes and they do not have an inversion symmetry [16]. The substrate has a big influence on the growth mode and the final physical and chemical properties, determining the surface morphology, polarity, crystal orientation, composition and elastic strains [17]. Because the high-end applications of GaN are currently expanding [18], the aim of the present study is to obtain semi-insulating GaN thin-films by RF magnetron sputtering on Al₂O₃ substrates. The deposited films will present some intentional and unintentional impurities incorporated which should hold promises of increasing the crystalline quality of the deposited material. The intentional impurity refers to a low amount of Ga₂O₃ found in the target material, which is bound to be transferred to the substrate and the unintentional impurities are related to the different drift speeds of Ga, O and N species in the plasma profile that can lead to a composition gradient along the surface of the substrate. Thus, the main goal of this study is to establish the structure and the morphology of gallium nitride films grown on Al₂O₃ substrates. For microstructural analysis X-ray diffraction is used and atomic force microscopy and scanning electron microscopy are used for surface morphology characterization.

2. Experimental results

2.1. Sample preparation

Growth of GaN thin films is performed by radio frequency (RF) sputtering using a UHV (ultra-high vacuum) deposition cluster. Before deposition, the Al₂O₃ substrates are cleaned with ethanol, fixed with Ag paste on the sample holder, kept on a hot plate for 20 minutes at 100°C to evaporate the solvent from the Ag paste. To dispose the possible remaining impurities, the samples are annealed for 2-3 hours at 300-550°C under vacuum ($\sim 10^{-6}$ mbar). The deposition conditions consist in a working pressure (P_d) of 1 Pa, made up of a mixture of Ar/N₂ in the plasma volume. The substrate is set to a distance (d_{ts}) of 20 cm from the target and its temperature is set to 550°C. The magnetron works under a power (P_s) of 100 W, and a presputtering, for cleaning the target, was done for 30 minutes with the shutter closed. Further, the GaN target is subjected to RF-magnetron sputtering for 90 minutes. After deposition, the sample heating and the gas are turned off, and the samples are cooled down (~ 10 °C/min) inside the deposition chamber to room temperature.

2.2. Sample characterization

2.2.1. Surface morphology

Ex-situ atomic force microscopy characterization (surface morphology, topography, roughness), performed in the semi-contact mode, shows that the growth type is Stranski-Krastanov, *i.e.* 2D- islands growth for the very thin films (~ 10 nm) with transition to a 3D growth for the thicker films (~ 34 nm). AFM images for thin/thick GaN films grown on Al₂O₃ substrate are represented in Fig. 1.

The increase in the root mean square (rms) roughness from ~ 0.45 nm for the thinner films to ~ 0.93 nm for the thicker films, is consistent with Stranski-Krastanov growth. It is assumed that at the initial stage of the growth, the film follows a 2D growth mode (coherent growth), with unit-cell height islands and, after relaxation, a transition to a 3D growth mode (2D to 3D transition) takes place, the film morphology indicating a 3D islands structure due to the lattice mismatch between the substrate and film.

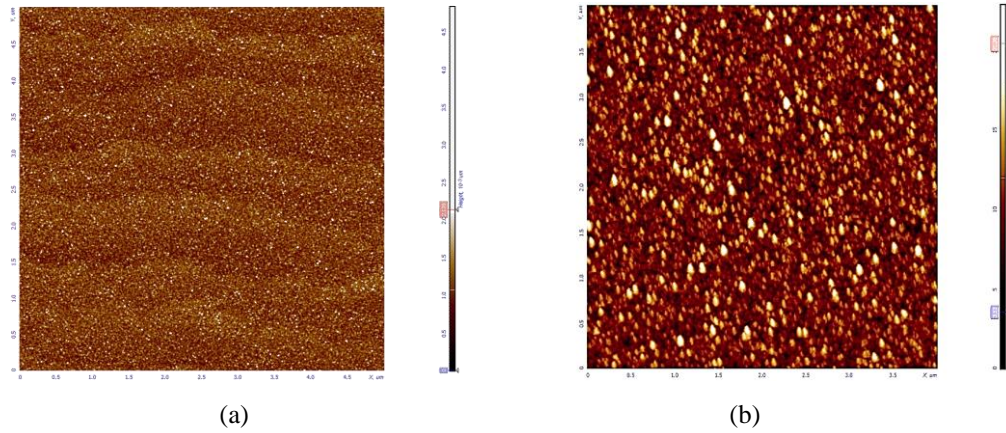


Fig. 1. AFM images of the films with different thickness grown on Al_2O_3 substrate (a) thin film (~ 9 nm) – the terraced structure of the substrate is still visible, (b) thick film (~ 34 nm) – the film shows a 3D islands structure

The scanning electron microscopy analysis is done in high vacuum. A flat, smooth, uniform surface is observed, free of cracks and with homogeneous morphology at few micrometers scale (see Fig. 2.).

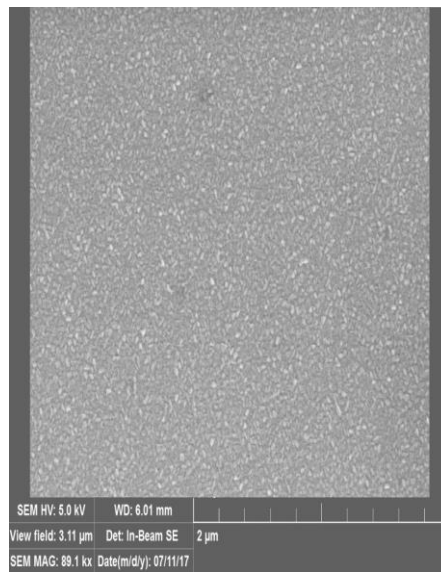


Fig. 2. Secondary electron SEM image of the ~ 34 nm film deposited on Al_2O_3 substrate

2.2.2. XRD structural analysis

Out-of-plane 2theta-omega scans are performed in order to determine the structure of the films (see Fig. 3). The films show a mixed phase composition, with two preferred orientations, one belonging to the GaN hexagonal crystal structure (PDF4+ 00-050-0792 [19]) (100), as the main phase (the first peak in the 2theta-omega scans in Fig. 3), and (104), as the minor phase (the second, lower intensity peak in the 2theta-omega scans in Fig. 3) belonging to the Ga_2O_3 rhombohedral crystal structure (PDF4+ 00-006-0503 [20]). The as-deposited films correlate with the chemical composition of the target which contains some amounts of Ga_2O_3 . Also, due to a lattice parameter similarity of Al_2O_3 ($c=12.993\text{\AA}$) (PDF4+ 00-046-1212 [21]) and Ga_2O_3 ($c=13.429\text{\AA}$), at the initial stage of the growth the more thermodynamically stable phase that is deposited is the oxide rather than the nitride, as indicates also the X-ray reflectivity data.

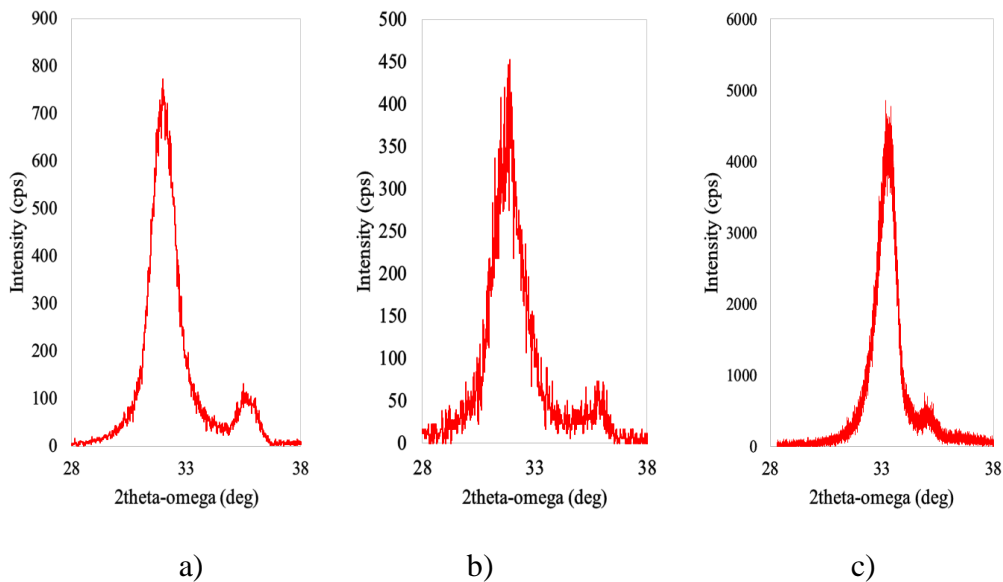
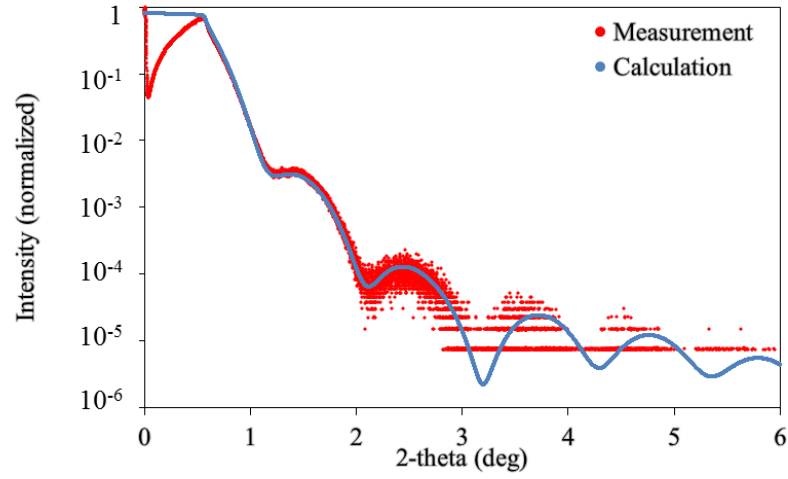
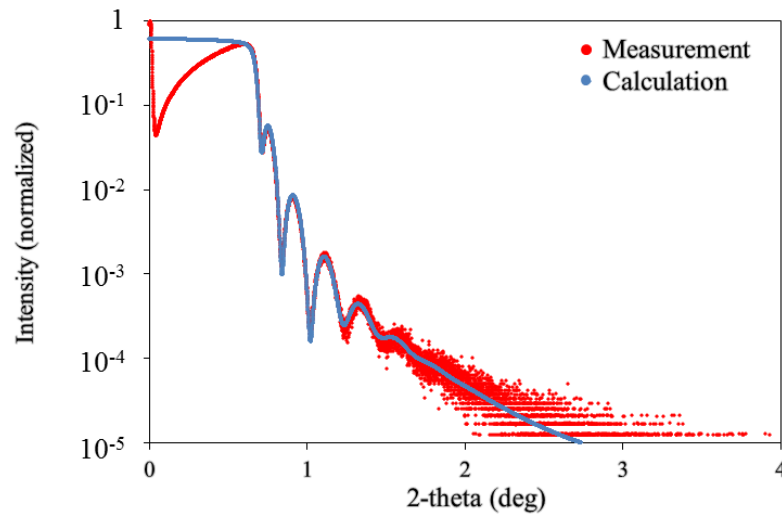


Fig 3. XRD 2theta-omega scan for the film grown on Al_2O_3 substrate; a) $\sim 9\text{nm}$ film, b) $\sim 11\text{nm}$ film, c) $\sim 34\text{ nm}$ film

X-ray reflectivity is used to determine several parameters of the films, such as thickness, the layered structure, the density, and roughness information. Some of the results are shown in Fig. 4, while in Table 1 are given the physical properties of the films as determined from reflectivity measurements. The reflectivity data simulate well when the substrate/oxide/nitride layer sequence is considered.



(a)



(b)

Fig.4 XRD reflectivity measurement of (a) ~ 9 nm thick films and (b) ~ 34 nm thick film grown on Al_2O_3 (0001)

Table 1.

Properties of the films resulted from X-ray reflectivity measurements

Substrate	Al_2O_3		
Total film thickness (nm) (GaN+ Ga_2O_3)	8.8(2)	11.1(2)	33.8(6)
GaN thickness (nm)	4.2(2)	7.2(2)	30.3(6)

GaN density (g/cm ³)	6.4(2)	6.3(2)	5.9(2)
GaN roughness (nm)	0.7(3)	0.6(3)	2.7(2)

3. Conclusions

Layers of variable thickness of gallium nitride/oxide mixtures are synthesized on Al₂O₃ substrates. The layer thickness ranges between ~9 nm to ~34 nm. From AFM measurements, the root mean square roughness is obtained; the suggested growth mode is Stranski-Krastanov (2D islands formation at the initial stage of the growth, followed by a 3D islands growth after film relaxation). The XRD analysis shows that the films had grown as an intermixing of hexagonal GaN (100) phase and rhombohedral Ga₂O₃ (104) phase.

In summary, the as-deposited films correlate with the chemical composition of the GaN target which contains some amounts of Ga₂O₃. Experiments with a single crystal GaN target are in progress, in order to obtain single-phase GaN films on different substrates (i.e., Si, Al₂O₃, ZnO, etc.)

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