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Properties of MOVPE GaN grown on ZnO deposited on Si(001) and Si(111) substrates

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ABSTRACT

GaN/ZnO layers were grown on p-type Si(001) and Si(111) substrates to develop preparation techniques of multi-material devices for optoelectronic and sensor applications.

At the first stage the thin, polycrystalline, zinc oxide (ZnO) buffer layers were deposited on p-type Si substrates by radio frequency (RF) sputtering techniques. The planar RF sputtering Perkin Elmer 2400/ 8L diode system equipped with hot-pressed ceramic ZnOAl target (99.99%purity) was employed. The sputtering power was 300 and 150 W, accordingly thicknesses of ZnO:Al films were 280 and 250 nm.

At the next stage the GaN layers were grown on ZnO/Si substrates in an atmospheric pressure, single wafer, horizontal flow metalorganic vapor phase epitaxy (MOVPE) system. The low frequency (40 kHz) inductive heating method was used to raise the temperature of graphite susceptor up to 1030 °C. Trimethylgallium (TMGa), trimethylaluminium (TMAI) and ammonia (NH₃) were used with H₂ carrier gas. The new type of temperature graded nitride multi-layers (NMLs) were applied and the multi-stage growth process was developed to prevent the ZnO layer decomposition during exposure to NH₃ and H₂ at high temperature.

Atomic force microscopy (AFM) and X-ray diffractometry (XRD) were applied to study the morphology and structural properties of the ZnO and GaN layers. The experimental conditions were selected for successful integration of high temperature gallium nitride (HT-GaN) layers with the ZnO films on Si substrate.

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CRYSTAL GROWTH

1. Introduction

Recently, GaN and ZnO both wide-bandgap semiconductors, have attracted considerable interest because of their potential application for optoelectronic and sensor applications due to its direct wide-bandgap of 3.4 eV and nearly the same lattice constants of 3.25 and 3.16 Å, respectively. The similarity of GaN and ZnO in terms of energy gap, crystallographic structure and high thermal stability make them attractive candidates for heterostructures forming. GaN was grown epitaxialy on ZnO substrates [1,2] and ZnO layers were deposited on sapphire substrates by different techniques such as atomic layer epitaxy (ALE), pulsed laser deposition (PLD) or radio frequency (RF) sputtering but only very few groups have studied the growth of GaN on ZnO layer grown on silicon substrates [3–6].

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The deposition of high quality GaN and ZnO on (111)Si and especially (001)Si substrates is essential for the full AIIIBV and Si material system integration which allowed the realization of chip, large area sensors and biosensors [7].

Although the lattice and thermal mismatches between ZnO and GaN are only 2% and 14%, respectively, the lattice and thermal misfits between Si and GaN are as large as 20% and 56%. The large difference in thermal expansion coefficients causes the cracking of GaN grown on Si substrate when the thickness of epitaxial layer exceeds 1 μ m. In order to avoid such cracks buffer layers have to be inserted between Si substrate and GaN layer. ZnO and AlN layers separately have proved their effectiveness in inhibiting cracks in GaN/Si epitaxy. To our knowledge no one has studied the growth behavior of GaN–AlN–ZnO–Si system.

The other serious problem of the growth of GaN and ZnO on silicon is the thermal stability of ZnO layer during the metalorganic vapor phase epitaxy (MOVPE) process. It was observed earlier [8,9] that the exposure of ZnO surface to hydrogen (H_2) and

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ammonia (NH₃) at temperatures above 600 °C results in the decomposition of ZnO and subsequent poor nucleation of GaN. It proved that for fabrication of the high quality MOVPE GaN/ZnO/Si layer the growth process has to contain stages which allow the technologist to avoid the thermal decomposition of ZnO layers during the growth.

In this paper, the properties and the growth process of MOVPE GaN on RF sputtered ZnO layers deposited on Si(001) and Si(111) substrates were studied. The new type of temperature graded GaN and AlN multi-layers were elaborated and the multi-stage growth process was developed which allowed the integration of GaN and ZnO layer on silicon substrate.

2. Experimental details

The polycrystalline ZnO:Al layers were deposited by RF sputtering technique on p-Si $(p \sim 1 \times 10^{18} \text{ cm}^{-3})$ substrate from hot-pressed ceramic ZnO:Al₂O₃ target in Ar atmosphere. Presputtering for 120 min was applied to clean the target. The sputtering power was changed in the range of 300-150 W and accordingly the thicknesses of ZnO:Al layers were in the range of 280-250 nm. The structural and electrical properties of processed ZnO:Al layers were investigated. Films deposited by RF sputtering show conductive ZnO:Al n-type. The surface morphology of ZnO layers exhibit strong dependence on deposition conditions. After deposition all ZnO/Si templates were annealed by rapid thermal annealing method in the temperature range of 400–500 °C using N₂ atmosphere for 3 min. The improvement of ZnO layers optical and electrical characteristics of the ZnO/Si templates was observed [10]. In Fig. 1a and b the surface morphology atomic force microscopy (AFM) images of ZnO:Al films obtained for different sputtering powers (300 and 150 W) are shown. The most homogenous surface morphology and grain formation with peakto-peak value up to \sim 33 nm was revealed for 300 W sputtering power (Fig. 1a). For 150W sputtering power the layer with columnar structure was obtained and the roughness increased up to 55 nm (Fig. 1b).

It was observed that the crystallographic quality of the sputtered ZnO has been improved with the layer thickness increase. All samples have more or less visible texture with the dominant ZnO (00 1) orientation. For layers deposited at 300 W of sputtering power at X-ray diffraction spectra (Fig. 1c and d) taken in $\Theta/2\Theta$ -mode dominates only ZnO (00 1) peak at 2 Θ angle of 16.57° while the layer sputtered at 150 W was polycrystalline with ZnO grains of different orientations: (00 1) and (100) at 2 Θ angle of 16.57° and 32.83°, respectively. The orientation of the Si substrate showed no influence on crystallographic quality of the ZnO layer which depends only on the condition of the sputtering process, mainly on the value of the RF power during deposition.

Before loading into the MOVPE reactor the ZnO/Si templates were degreased in sequential ultrasonic baths of trichloroethylene, acetone and isopropyl alcohol for 30 min at 50 °C and blow dried with nitrogen. At the next stages the samples were loaded into an atmospheric pressure, single wafer, horizontal flow MOVPE system. The low frequency (40 kHz) inductive heating method was used to raise the temperature of graphite susceptor up to 1030 °C. Trimethylaluminium (TMAl), trimethylgallium (TMGa) and ammonia (NH₃) were used with H₂ carrier gas as a precursors. The MOVPE process started with a thermal baking of the ZnO/Si templates in H₂ for 10 min at 250 °C, next, the thermal baking has been continued at H₂ and NH₃ mixture for 10 min at 450 °C. Then, the four types of nitride multi-layers (NMLs) were deposited at low and medium temperature, classified as A (temperature graded AIN/ LT-GaN/LT-AIN/temperature graded GaN, 170 nm thick), B (temperature graded GaN/LT-AIN/LT-GaN/temperature graded GaN, 170 nm thick), C (temperature graded AlN/temperature graded GaN, 100 nm thick), D (LT-GaN/temperature graded GaN, 170 nm thick). These NML were inserted between high temperature gallium nitride (HT-GaN) and ZnO/Si for strain relief purpose and complete covering of the ZnO layer to prevent its decomposition during growth of HT-GaN. The temperature graded multi-layers of GaN and AlN were applied because in our early study of GaN growth on a- and c-oriented sapphire we experimentally verified their effectiveness for improvement of the quality of GaN subsequent epitaxial layer [11,12].



Fig. 1. AFM images (a, b) and XRD $\Theta/2\Theta$ -scans patterns (b, c) of ZnO/Si structures deposited at different RF sputtering powers 300 W (a, c) and 150 W (b, d), respectively. The substrate orientations were (111)Si (a, c) and (001)Si (b, d).

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