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Study of Al diffusion in GaN during metal organic vapor phase epitaxy of AlGaN/GaN and AlN/GaN structures

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ABSTRACT

$Al_xGa_{1-x}N$/GaN hetero-structures were grown on SiN-treated (00.1) sapphire substrate by atmospheric pressure metalorganic vapor phase epitaxy (AP-MOVPE). Characterization of the grown structures was performed in-situ by laser reflectometry and ex-situ by secondary ion mass spectrometry (SIMS) measurements. Al SIMS profile showed some tailing to GaN layers which is associated to Al diffusion. The trimethylaluminium (TMA) effects on the growth rate, Al composition and Al diffusion coefficient were discussed. Al diffusion coefficients (D_{Al}) into GaN were calculated. The results suggest that Al diffuses faster near the AlN/GaN interface.

1. Introduction

III–V nitride materials have attracted great attention as a promising system for applications in high-power and high-temperature electronics and opto-electronic devices operative in UV and visible wavelengths [1–2]. With a direct band gap ranging from 3.41 eV to 6.2 eV for GaN and AlN respectively, the alloy system offers unique opportunity for device design. $Al_xGa_{1-x}N$ alloys are under investigation over the past few years due to their potential in AlGaN /GaN heterostructures. Devices employing these heterostructures have recently been fabricated including; UV blue laser diodes (LDs), UV photodetectors and high electron mobility transistors (HEMTs) [3–5]. It is well known that the properties of AlGaN/GaN interface strongly affect the performance of these devices [6,7]. Thus, structure design and control of the specific Al distribution at the interfaces are extremely important. The significant diffusion of Al in III-nitride semiconductors and high Al content in $Al_xGa_{1-x}N$ layer causes an Al concentration profile with a long tail and results in an imperfect AlGaN/GaN interface that degrades the performance of devices [7,8]. The Al diffusion governs the final quality of the interface which takes on a characteristically asymmetric shape. Cai et al. [8] have reported that the diffusion of Al element across the interface at high growth temperature intrinsically exists and is difficult to cancel. Consequently, understanding the Al incorporation efficiency and the redistribution behavior of Al atoms during MOVPE growth is necessary. Particularly, diffusion behavior is an important issue that helps to well control the AlGaN/GaN interface. Until now, investigations on the Al diffusion in GaN have dealt with the Al interdiffusion near the GaN-sapphire interface [9–11]. However, improving interfacial abruptness amidst high temperature diffusion is an even greater challenge in the fabrication of nitride based devices [8,12]. In this paper, we report on the results of Al profile across the $Al_xGa_{1-x}N$/GaN and AlN/GaN interfaces by SIMS measurements. From these profiles measurements, we show the layer thickness and Al amount effects on the Al diffusion coefficient.

2. Experimental details

$Al_xGa_{1-x}N$/GaN hetero-structures were grown on c-plane sapphire by AP-MOVPE in a vertical reactor. Trimethylgallium (TMG), trimethylaluminium (TMA), and ammonia (NH_3), were used as precursors for Ga, Al and N, respectively. A mixture of hydrogen (H_2) and nitrogen (N_2) were used as carrier gas. The growth was initiated with a standard sapphire nitridation followed by a SiN thin mask deposition. Then, a GaN buffer layer (BL) with thickness of about 30 nm was deposited at 600 °C followed by a GaN template layer grown at high temperature (1100 °C). This structure was used as pseudo-substrate for the growth of $Al_xGa_{1-x}N$ layers. Details of the growth procedure were reported elsewhere [13].

Three hetero-structures labeled A, B and C composed of alternating $Al_xGa_{1-x}N$ and GaN layers were grown at 1100 °C. For

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sample (A), two $Al_xGa_{1-x}N$ spikes (S_1, S_2) separated by GaN layer were epitaxially grown using 3 slm (standard liter per minute) of NH_3, 40 μmol/min of TMG and 12 μmol/min of TMA. This structure was terminated by an AlN cap layer. For sample (B), three AlGaN spikes were grown under the same growth conditions as sample A, but with different TMA flow rates, 7 μmol/min (S_3), 12 μmol/min (S_4) and 19 μmol/min (S_5) spaced and capped with GaN layers. The sample C consisted of GaN/AlN/GaN hetero-structure. The growth monitored in-situ at normal incidence by He-Ne laser reflectometry (λ=632.8 nm). Calibrated SIMS analysis was used to determine the Al depth profiles along the growth direction of the three samples A, B and C. For a high depth resolution, SIMS analysis was carried using an impact energy of 3 keV for the (Cs^+) primary ion beam and monitoring positive secondary ions. Absolute Al concentration was obtained by comparison with a reference sample. Relative sensitivity factors (RSFs) were used for quantitative calibration of secondary ion intensities RSF= $C_M/(I_M/I_{ref})$; C_M and I_M are concentration and secondary ion intensity of element M, respectively, and I_{ref} is the secondary ion intensity of a reference element. The RSF calibration coefficient of Al in GaN was derived from a reference sample. The photoluminescence (PL) measurements were made at room temperature, using the 325 nm line of He–Cd Laser. Atomic force microscopy (AFM), on 5 × 5 μm scale, was used to determine the surface morphology.

3. Results and discussion

Fig.1(a) illustrates the in-situ reflectivity signal recorded during the growth of sample A. Firstly 0.5 μm GaN layer was deposited at 1100 °C. When the reflectivity signal showed a bi-dimensional growth mode, the first $Al_xGa_{1-x}N$ layer was grown by introducing 12 μmol/min of TMA flow rate into the reactor. After 10 min, the TMA was switched OFF, and a GaN layer was grown. This first AlGaN/GaN sequence was followed by a second one and capped with an AlN layer. We observed a damping of the amplitude of the oscillations with deposition time. This could be explained by optical absorption below band gap and/or optical scattering of a non-planar surface [14]. The turn ON and OFF of TMA was respectively accompanied by an increase and a decrease of reflectivity period oscillations. This is related to AlGaN growth rate reduction with respect to that of the GaN layer one. The growth rate (V_g) is deduced from the reflectivity oscillation period [15,16].

In order to check the Al solid composition, calibrated SIMS analysis was done. Al and Ga SIMS profiles for sample A were shown in Fig. 1(b). The two intentionally sandwiched AlGaN spikes (S_1 and S_2) were identified. We found 1.9% and 2.3% of Al solid composition in S_1 and S_2 respectively. The corresponding growth rates of the first and the second AlGaN layers are at about 1.5 and 1 μm/h respectively. These Al compositions are in agreement with those determined by photoluminescence and high resolution x-ray diffraction measurements of AlGaN layers grown at the same conditions (not shown here). The S1 spike was grown on an incompletely coalesced GaN template layer, compared to the S2 one. In our case, by using the SiN treatment growth method, the coalescence (transition from 3D to 2D growth mode) was achieved at about 1 μm of thickness. By using the same epitaxial conditions as sample A, the growth of two GaN templates layers was interrupted at the same coalescence degree as those used for the deposition of the two AlGaN layers. AFM images of the two GaN templates were shown in Fig. 2. One can note that the first spike S1 was grown on a not fully coalesced GaN layer as shown in Fig. 2(a). However the second spike was grown on a fully coalesced GaN template (Fig. 2 (b)). The near interface film/substrate region is characterized by high defect density. After the coalescence, the film surface morphology is uniform [17]. In a previous work [18], we have reported that AlGaN layers grown on GaN templates with different coalescence degree showed a slight variation of about 0.5% in Al solid composition. Furthermore, it should be noted that the first grown spike S1 was annealed for a longer time compared to the second

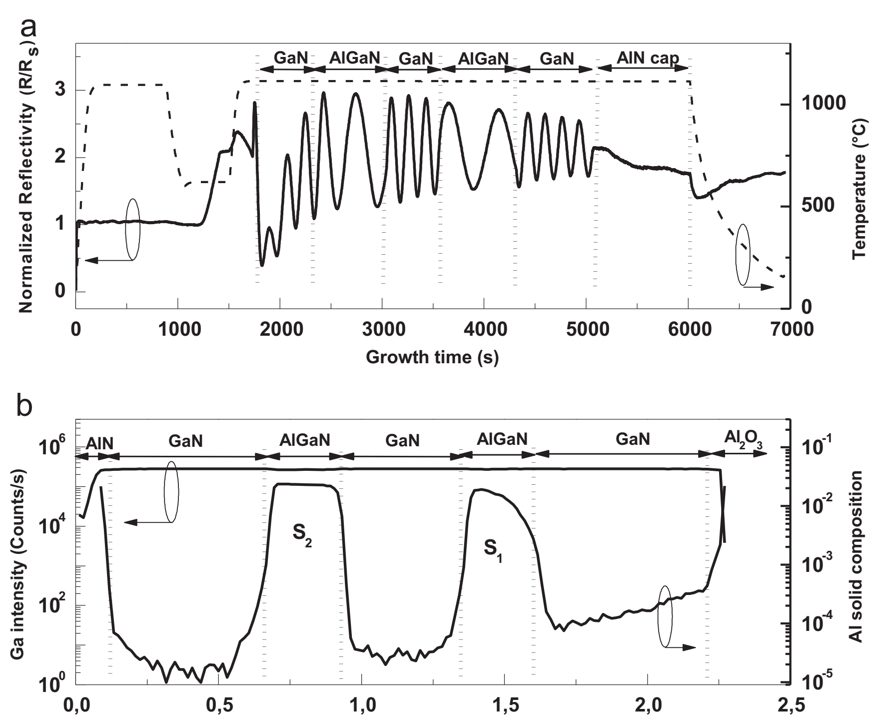

Fig. 1. (a): In-situ reflectivity and growth temperature plots versus growth time recorded during the growth of sample A. The dashed lines show the change of reflectivity behavior from one layer to another. (b): SIMS profiles of Ga and Al solid composition in sample A. The flow rates of NH_3, TMG and TMA were respectively maintained at 3 slm, 40 μmol/min and 12 μmol/min.