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P5-7

Flat surfaces and interfaces in AlN/GaN heterostructures and superlattices grown by flow-rate modulation epitaxy

Masanobu Hiroki, and Naoki Kobayashi

NTT Basic Research Laboratories, NTT Corporation
3-1, Morinosato Wakamiya, Atsugi, Kanagawa 243-0198, Japan
Phone: +81-46-240-3354 Fax: +81-46-240-4729 E-mail: hiroki@will.brl.ntt.co.jp

1. Introduction

AlN/GaN or AlGaN/GaN heterostructures and superlattices (SLs) with high Al compositions have attracted much attention recently for applications to heterostructure field effect transistors [1], optical devices using the intersubband transition at the communication wavelength [2], and resonant tunneling diodes [3], due to their high band offset and large polarization effect. Flat heterointerfaces and control of the Al composition are key factors for those devices; however, there are difficulties in growth of flat and uniform AlN layers on GaN by metalorganic vapor phase epitaxy (MOVPE). The parasitic reaction of ammonia (NH_3) and trimethylaluminum (TMA) in vapor phase makes it difficult to control the Al compositions precisely. Usually high growth temperatures exceeding 1100°C are needed in order to grow directly high quality AlN on silicon carbide (SiC) [5,6] or sapphire substrates [7]. But such high temperature growth is not practical on GaN due to the surface etching problem.

Flow-rate modulation epitaxy (FME) is a growth method in which group III sources and group V sources are supplied alternately [8]. In this work, we applied FME for the fabrication of AlN/GaN heterostructures and compared the AlN flatness and Al composition control with those achieved when the sources are simultaneously supplied. The results indicate FME is an effective growth method for the fabrication of high quality AlN/GaN heterostructures and SLs with flat interfaces.

2. Experimental

AlN/GaN was grown by horizontal MOVPE. Growth pressure was 300 Torr. GaN template layers were grown on c-plane sapphire substrates. Trimethylgallium (TMG), TMA and NH_3 were used as the precursors for Ga, Al and N. In FME, one cycle of AlN growth consists of 1- or 2-sec supply of TMA and 1-sec of NH_3. There was no growth interruption between the TMA and NH_3 supply. The growth rate of AlN was estimated by in-situ optical reflection monitoring with a He-Ne laser (632.8nm). The Al compositions in AlGaN were measured by a 2θ-ω scan in x-ray diffraction (XRD). The surface morphologies of AlN on GaN were observed by atomic force microscopy (AFM).

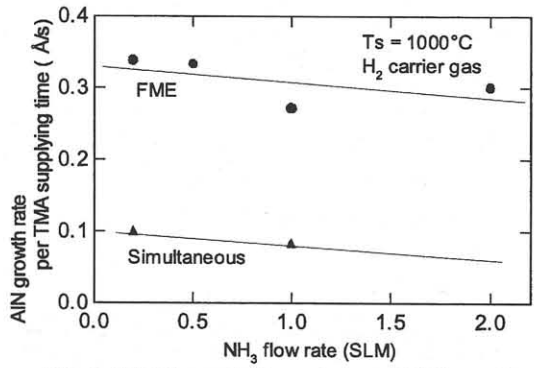

Fig. 1. NH_3 flow-rate dependence of AlN growth rate.

3. Results and discussion

Firstly, we compared the AlN growth rate between FME and the simultaneous supply. Figure 1 shows the NH_3 flow-rate dependence of the AlN growth rate at the constant TMA flow-rate of 1.2 μmol/s. The growth rate of AlN grown by FME was about 0.3 Å/s, which was three times higher than for the simultaneous supply. It is considered that the Al atoms are taken into solid phase from TMA more efficiently in FME, as a result of reducing the reaction between TMA and NH_3 in vapor phase.

The reduction of the reaction between TMA and NH_3 by FME was also confirmed in the control of Al compositions of AlGaN. Figure 2 shows the dependence of Al compositions on the TMA ratio in the vapor phase for the simultaneous supply with H_2 carrier gas, FME with H_2 carrier gas, and FME with N_2 carrier gas. Al compositions were saturated at about 40% by the simultaneous supply due to the reaction of TMA and NH_3. This is a commonly observed phenomenon in AlGaN MOVPE. On the other hand, the saturation of Al compositions was not observed at the FME growth with either carrier gas due to the reduction of the reaction between TMA and NH_3. However, the Al compositions are much higher with H_2 carrier gas at the same vapor phase ratio. It has been reported that H_2 gas etches the GaN surface [9]. So Ga atoms evaporate at the moment when group III is supplied without NH_3 flow. With N_2 carrier gas, Al compositions nearly equal the vapor phase ratio. The evaporation of Ga atoms was suppressed in N_2-FME. The higher growth rate of AlN and the good controllability of Al composition in AlGaN

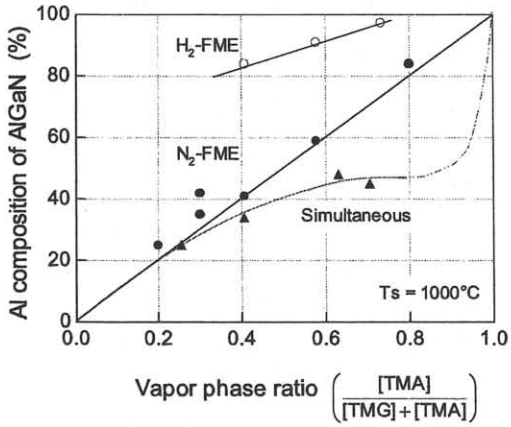

Fig. 2. Dependence of Al composition on the TMA ratio in the vapor phase

achieved in FME clarify that FME is suitable for the growth of AlN and AlGaN.

Next, the structural properties of FME-grown AlN/GaN heterostructures were studied. AlN was grown on GaN by FME and simultaneous supply at 1000°C. The thickness of AlN was fixed at about 3-4 nm, which is nearly the critical thickness of AlN on GaN [10]. Figure 3 shows AFM images of AlN surfaces. The surface of AlN grown by the simultaneous supply is rough, consisting of islands about 50nm in diameter. In contrast, the AlN by FME has a flat surface on which atomic monolayer steps can be seen. The island growth of thin AlN on GaN has already been reported for the simultaneous supply [11]. Three possible reasons for three-dimensional growth are the lattice mismatch of 2.4% between AlN and GaN, increases in V/III ratio due to the reaction between TMA and NH_3 and the short migration length of Al atoms due to the large bond strength of AlN. FME overcomes the disadvantages of simultaneous supply with respect to the second and the third reasons. FME suppresses increases in V/III ratio by reducing the reaction in the vapor phase. The monolayer steps seen on FME-AlN surfaces suggest a long migration length in FME growth.

AlN/GaN SLs were also fabricated. Satellite peaks up to the 4th order were clearly seen in XRD measurements for 10-periods of AlN (3nm)/GaN (22nm). This indicates abrupt heterointerfaces were formed by FME. Strained SLs could make dislocations bend laterally and thereby lead to a reduction of the threading dislocation [12]. GaN was grown on AlN/GaN SLs and the dislocation densities were estimated by the etch pit density (EPD). There was less of reduction of dislocation density in the case of simultaneous supply. This may be due to the existence of the pits on the surface, as observed in Fig. 3. Considering the pit density (1.2×10^9 cm^{-2}), the pits originate from dislocations. The pits relaxed the interface strain around the dislocations; the dislocations are not bent effectively and propagate easily

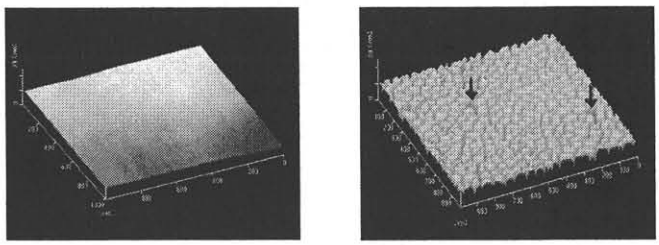

Fig. 3. 4nm-AlN surface morphologies on GaN observed by AFM. Arrows denote pits originating from the threading dislocations.

to the surface in the AlN/GaN SLs grown by simultaneous supply. In contrast, EPD was reduced to about 1/2 by FME; few pits are observed in the FME-AlN surface.

4. Conclusions

AlN/GaN heterostructures and SLs were grown by FME. The reaction between TMA and NH_3 were much reduced by FME. The growth rate became faster and the control of Al composition in AlGaN was much improved compared to the simultaneous supply. The surface of FME-grown AlN on GaN was flat. The AlN/GaN SLs grown by FME have abrupt and flat heterointerfaces and reduced dislocation density.

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