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In-situ real-time analysis on strain relaxation process in GaN growth on sapphire by RF-MBE

K. Xu[a,*], N. Yano[b], A.W. Jia[a,b], A. Yoshikawa[a,b], K. Takahashi[c]

[a] *Center for Frontier Electronics and Photonics, Chiba University, 1-33 Yayoi-cho, Inage-ku, Chiba 263-8522, Japan*
[b] *Department of Electronics and Mechanical Engineering, Chiba University, Japan*
[c] *Department of Media Science, Teikyo University of Science and Technology, Japan*

Abstract

In-situ and real-time monitoring on RF-MBE growth of GaN on sapphire substrate was carried out by using spectroscopic ellipsometry and reflection high-energy electron diffraction. Two relaxation processes were identified through the accompanying surface roughening in RF-MBE growth of GaN. The first relaxation of GaN on sapphire substrate happened through Stranski–Krastanov mode growth in the very initial stage of buffer. With further deposition, a smooth buffer layer was obtained and growth mode was changed to 2D. The second relaxation took place during the buffer layer temperature ramping process, and the relaxation temperature depended on the buffer layer growth temperature. After the second relaxation, the buffer layer became partly connected islands, as confirmed by AFM observation. These two relaxation processes were revealed to have important effects on epilayer quality.

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1. Introduction

III–V nitrides including GaN, AlN and InN have evoked great interest because of their promising prospects in applications to optoelectronic devices [1,2]. However, growth of high-quality III-nitride films is still a major problem in achieving the anticipated performances of the GaN-based devices, especially in the case of MBE growth.

MBE is thought to have several advantages over MOVPE. But from the viewpoint of material quality, MBE technique lagged behind and obtained only limited success in a few areas such as UV photodetectors and HFETs. With great efforts by many groups in recent years [3–7], much progress has been made in quality of films grown by MBE so that the quality of III-nitrides can be comparable to the best MOVPE-grown layers.

Many techniques have been developed to grow high-quality GaN by MOVPE, but they are not applicable in MBE growth of GaN. The difference of growth mechanism between MOVPE and MBE is not yet well elucidated. Much effort is still needed to understand MBE growth mechanisms well, such as how GaN is relaxed on sapphire substrate in MBE growth. In the present work, our

*Corresponding author. Fax: +81-43-290-3988.
E-mail address: xuke@vbl.chiba-u.ac.jp (K. Xu).

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experimental results of RF-MBE growth with in-situ monitor and control by using spectroscopic ellipsometry (SE) and reflection high-energy electron diffraction (RHEED) are reported. The basic process of RF-MBE growth of GaN is clarified, and several crucial issues in RF-MBE growth of GaN are discussed. A better understanding of the related growth mechanisms has been attempted to expand the success of MBE-grown GaN in applications to optoelectronic devices.

2. Experimental procedure

Growth of GaN epilayers was performed in an RF-MBE system. Ga source was supplied with Ga vapor beam flux from an effusion cell and N by RF-plasma N cell (EPI Uni-bulb). The construction of RF-MBE system is shown in Fig. 1. The base pressure under liquid nitrogen circulation is in the order of 10^{-11} Torr. The system was equipped with RHEED, SE, UHV-STM/AFM, and Coaxial Impact Collision Ion Scattering Spectroscopy (CAICISS). A CCD camera was used to monitor RHEED screen so that RHEED pattern and spot intensity could be recorded and analyzed by KSA400 software in real time. The SE system can provide the dynamic simulation and spectroscopic monitoring with the wavelength ranging from 245 to 727 nm. This integrated MBE system enables us to evaluate and control the epilayer growth in situ and in real time.

The growth sequence of GaN is as follows: nitridation of sapphire substrate at 200°C GaN for 40 min, buffer layer growth by migration-enhanced epitaxy (MEE) method at different temperatures ranging from 350°C to 720°C, annealing and epilayer growth at 820°C. The growth rate of epilayer can reach 0.5 μm/h with a nitrogen flow rate of 0.65 sccm. The epilayers were also characterized by high-resolution XRD, PL, and room-temperature Hall measurements.

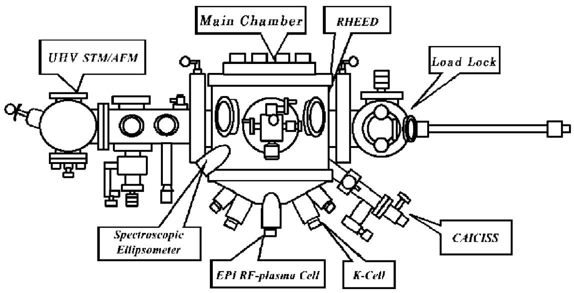

Fig. 1. Setting up of RF-MBE system.

3. Results and discussions

Fig. 2 shows the typical evolution of the real part $\langle \varepsilon_1 \rangle$ of pseudo-dielectric function during the whole growth process. The buffer layer growth temperature was 650°C.

With starting of GaN buffer layer growth on nitridated sapphire substrate, $\langle \varepsilon_1 \rangle$ increased gradually and reached a peak value (marked as A in Fig. 2) after around 5 monolayer (ML) growth. The decreasing of $\langle \varepsilon_1 \rangle$ after peak value at position A indicated S–K mode growth start of GaN buffer; this was the first relaxation of GaN on nitridated sapphire substrate. The longer the probing light wavelength is, the later $\langle \varepsilon_1 \rangle$ reaches a peak value. In this growth stage, SE signal variation depends on the competition between film thickness increase and surface roughening. The formed islands during the S–K mode growth were in nanometer or tens of nanometer scale, and the probing light with shorter wavelength is more sensitive to the surface roughening than longer wavelength light. Therefore, the peak A would appear later if the probing light wavelength was

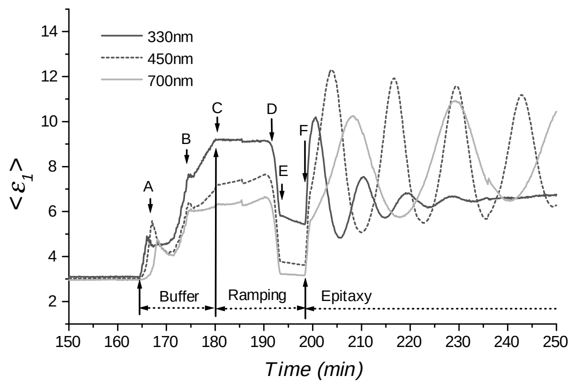

Fig. 2. Evolution of real part $\langle \varepsilon_1 \rangle$ of pseudo-dielectric function during GaN growth.

longer. The accompanying surface roughening was further confirmed by RHEED and AFM observation, as will be shown later. With further growth of the buffer layer, position B marked the slope change for SE signal increasing rate. Before position B, SE signal increased with higher rate due to the effect of surface smoothing and film thickness increasing; after position B, only thickness mainly contributed to the SE signal increasing. Buffer layer growth was stopped at position C corresponding to a thickness of 20–30 nm.

Fig. 3 shows the recorded RHEED patterns during the initial growth stage of the same buffer layer as shown in Fig. 2. At 0 min, the streaky pattern is for AlN [1 $\bar{1}$ 0 0]. AlN layer was formed during nitridation. After 1 min growth of buffer layer, a very streaky pattern from GaN layer was observed. The RHHED pattern at 2 min was recorded at left side of the peak position A in Fig. 2, the one at 3 min was taken at the right side of the peak A. The spottiest pattern appeared at 6 min, which was corresponding to the valley between A and B in Fig. 2, The surface morphology after 6 min growth is shown in Fig. 4(a); the size of self-organized dots is about tens of nanometers. In this stage, GaN buffer was grown through S–K mode, and relaxed by forming the nanometer-sized dots. With further deposition of GaN, RHEED pattern became streaky again after 11 min; this point corresponds to position B in Fig. 2. Fig. 4(b) shows the buffer layer morphology after 15 min growth. The surface became very smooth. The island size was around 200 nm.

Figs. 2–4 show the evolution of SE signal and RHEED pattern and surface morphology of the buffer layer grown at 650°C. The dependence of SE signal evolution on buffer layer growth temperature was also observed. We found that the first relaxation through S–K growth mode depended on the buffer layer growth temperature. S–K growth mode took place later and the peak was higher for the buffer layers grown at 350°C compared to those grown at 650°C. As a common point of the buffer layers grown from 350°C to 650°C, we always observed that the S–K mode growth took place after several MLs deposition. But for a higher buffer layer temperature, the growth behavior is very different. Fig. 5 shows the

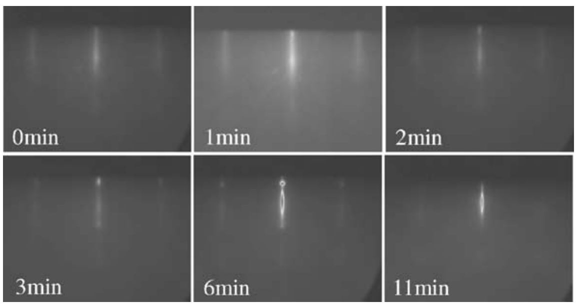

Fig. 3. RHEED patterns of GaN buffer layer at different growth stages, recorded along [1 $\bar{1}$ 0 0]. Nitridated sapphire substrate at 0 min is from AlN; streaky pattern from GaN at 1 min; starting to become spotty at 2 min; becoming spotty at 3 min; most spotty at 6 min; becoming streaky again after 11 min growth.

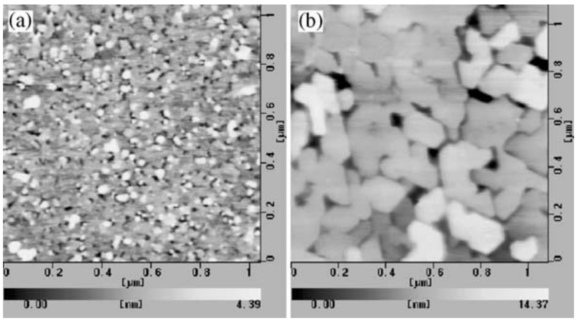

Fig. 4. AFM images of GaN buffer layers after 6 min growth (a), and after 15 min growth

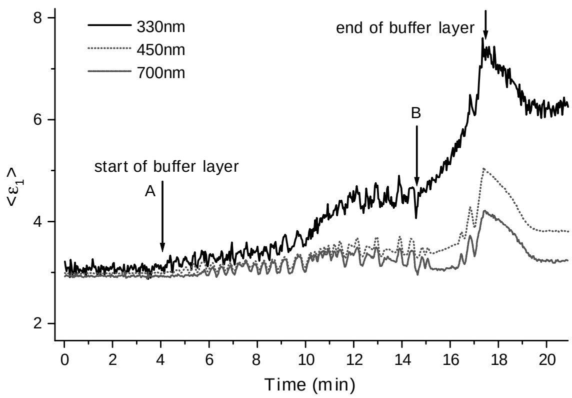

Fig. 5. SE signal $\langle \varepsilon_1 \rangle$ evolution during buffer layer growth at 720°C, the relaxation occurred simultaneously.

SE signal evolution of the buffer layer grown at 720°C. The deposited GaN was relaxed immediately, and the S–K mode dominated the growth from the buffer layer start point A to position B. Another different point is that, as can be seen in Fig. 5, $\langle \varepsilon_1 \rangle$ decreased quickly when the buffer layer growth was stopped, indicating that the surface became rough immediately.

The relaxation in the initial growth stage was revealed to play a very important role in RF-MBE growth of GaN. Because of the lattice mismatch between GaN and the nitridated sapphire substrate, the self-organized dots would relax themselves by crystallographic rotation and/or tilt. With further growth, when these dots meet together, dislocations would be generated to accommodate the differences of crystallographic orientation. We thought this was the main origin of the high threading dislocation density in RF-MBE-grown samples.

The second relaxation took place during buffer layer temperature ramping process, marked by a sharp drop of $\langle \varepsilon_1 \rangle$ in region D–E (Fig. 2). The corresponding temperature range was between 740°C and 760°C for the buffer layer grown at 650°C. For the buffer layer grown at 350°C, this second roughening took place between 720°C and 740°C. We have seen in Fig. 5, for the buffer grown at 720°C, the second relaxation took place immediately when buffer growth was stopped. The RHEED pattern corresponding to region D–E in Fig. 2 was changed from a streaky one to a spotty one with V-shaped wings, indicating that the surface became rough and morphology was featured by faceted islands. We can easily imagine that the surface roughening will occur preferably in the high-defect regions. In the further ramping process after surface roughening, threading dislocations in buffer layer may be bent to lie in the basal plane or vanished due to the surface mass transport and evaporation. Therefore, this is an important process for threading dislocation reducing.

When epilayer growth was started at position F, $\langle \varepsilon_1 \rangle$ for three wavelengths increased simultaneously with a steep slope, indicating that pseudo-2D growth occurred immediately. The subsequent oscillation of $\langle \varepsilon_1 \rangle$ originated from interference with film thickness increasing. The oscillation of $\langle \varepsilon_1 \rangle$ at 330 nm wavelength was flattened after epilayer thickness was more than about 300 nm due to adsorption.

Buffer layer growth temperature has remarkable effects on the epilayer crystal quality. When the buffer layer was grown at 350°C, FWHM of X-ray (0 0 2) reflection rocking curve could be as narrow as 35arcsec but that of (1 0 2) reflection was nearly 2000arcsec. AFM observation showed that the epilayer morphology was featured with 200 nm-diameter grain. The buffer layer grown at 650°C provided the best epilayer. There was no difference in epilayer surface morphologies for the samples with 650°C and 700°C buffer layer but FWHM of (1 0 2) was broadened. This was attributed to the relaxation process of the buffer layer during the ramping.

As far as we know, it is the first time that the S–K mode growth was noticed in the initial growth stage of GaN buffer layer on nitridated sapphire substrate. These two relaxation processes must be fully recognized to grow high-quality GaN epilayer by MBE.

4. Conclusions

In-situ monitored and controlled growth of GaN has been carried out to investigate the RF-MBE growth behavior. Two relaxation processes were identified in the very initial buffer layer growth stage and buffer layer temperature ramping process. The first relaxation was accomplished by S–K growth mode after several ML of GaN deposition, and self-organized quantum dots were formed during this stage. The relative crystallographic orientation differences of these dots were thought to be the origin of high threading defects density in MBE-grown epilayer. The second relaxation took place through surface roughening when the buffer layer temperature was ramped up. We suggested that, during the second relaxation and the latter annealing process, some threading dislocations might vanish or be bent into the basal plane, therefore epilayer quality could be improved by controlling these processes. Pseudo-2D growth was predominant in RF-MBE growth of

GaN. High-quality GaN epilayer was obtained based on the in-situ and real-time monitoring of GaN growth behavior and relaxation process by SE and RHEED.

Acknowledgements

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