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Measurement of X-ray CTR Signals from GaN/GaInN/GaN at High Temperatures Using Newly Developed Measurement System

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A combined system of laboratory X-ray CTR scattering measurement and MOVPE growth facility was set up and CTR measurement on GaN/GaInN/GaN heterostructure was conducted at room temperature and high temperatures up to 1000 ºC. Clear CTR signals and composition profiles were obtained even at 1000 ºC.

Key words: CTR, MOVPE, Laboratory X-ray, GaN/GaInN/GaN, High temperature

1. INTRODUCTION

In our previous papers, we reported the design and setup of the laboratory level X-ray CTR (crystal truncation rod) scattering measurement system and successful measurement of the CTR signals from InP/GaInAs/InP and GaN/GaInN/GaN heterostructures at room temperature within 100 min. The improvements made were to use a multi-layered focusing mirror and an asymmetry-cut double-crystal to collect and squeeze the X-rays from the rotating target X-ray source and to use slits between the sample and the detector to avoid stray X-rays [1-2].

Our intention is to investigate the growth process of, for example GaN/GaInN/GaN, using the monolayer-sensitive X-ray CTR scattering. The MOVPE (metalorganic vapor phase epitaxy) reactor with two Be windows was installed to the laboratory X-ray system [1].

In this paper, we report the proper operation of the measurement system with the reactor installed and the X-ray CTR scattering measurements on the GaN/GaInN/GaN heterostructure at as high as 1000ºC.

2. REACTOR AND STAGE

Though a picture of the X-ray system and installed reactor was shown in our previous paper [1], some details of the reactor structure and the XYZ-stage are described here. Fig. 1 shows the side view and front

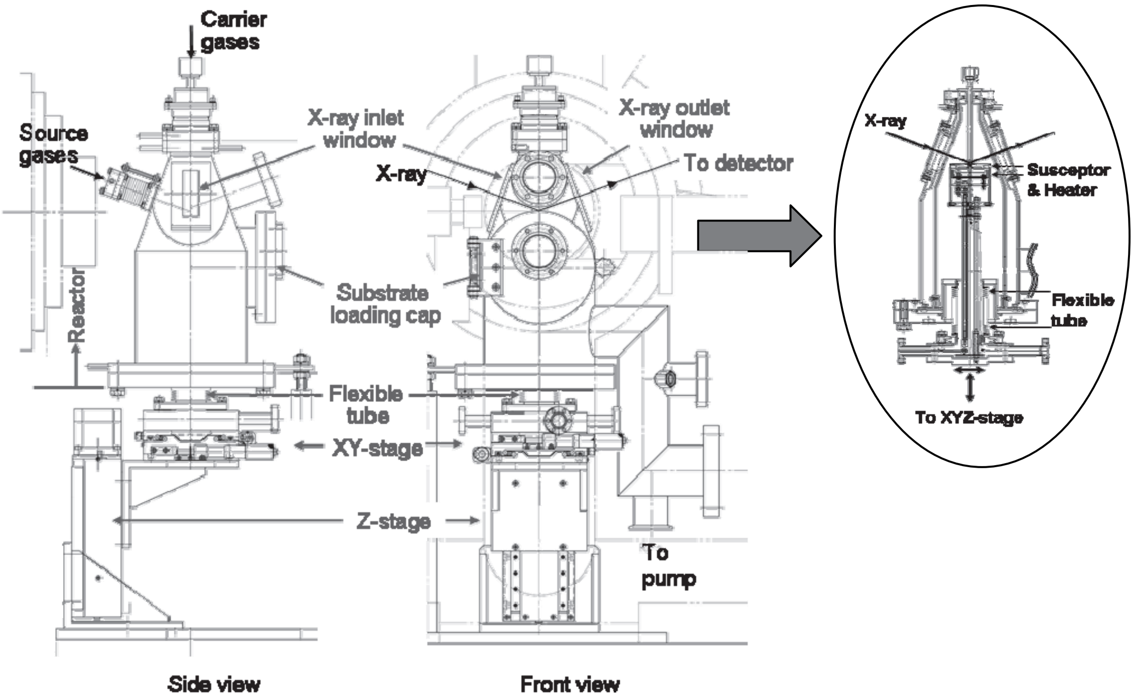

Fig. 1 Side view and front view of reactor and XYZ-stage for positioning the susceptor surface to the X-rays, while the reactor is fixed. Be windows are used for the X-ray path through the reactor. The inset is to show the susceptor and heater inside the reactor.

view of the reactor and stage. The reactor part is fixed at a proper position to the X-ray source and detector and there is no freedom for the reactor to be adjusted because pipes for gas introduction and tubes for pumping are fixed to reactor.

To adjust the sample position against the incoming and outgoing X-rays, an XYZ-stage was set to move the susceptor and heater part through a flange connected by a flexible tube to the reactor as shown in the inset of Fig. 1. The Z-stage is essential for sample positioning and is tough enough to support the XY-stage from the pulling-up force when the reactor was pumped to vacuum. The thickness of the Be window was 0.3mm. The intensity lowered to 85% of the original one due to the two Be windows.

3. HIGH TEMPERATURE MEASUREMENTS

3.1 S/B ratio

First of all, it is well anticipated that the thermal diffuse-scattering (B: background) increases and the CTR scattering intensity (S: signal) decreases at higher temperatures. To find whether the S/B ratio is high enough to measure the CTR scattering profile at higher temperatures, we measured the temperature dependence of the CTR peak intensity (at around 1.95 in Fig. 4) and thermal diffuse-scattering intensity using the GaN/GaInN/GaN wafer on sapphire substrate as shown in Fig. 2. In the temperature-change process it was necessary to wait for about 50 min until the susceptor and heater expansions were stabilized. The sample height was adjusted by the Z-stage.

The temperature dependence of the S/B ratio is shown in Fig. 3. The experimental data are shown by (○). The calculation of S was made using a standard diffraction intensity where the Debye-Waller factor

$$S \propto \exp(-2M) \qquad (1)$$

is the most temperature-dependent. M is a function of Θ_D/T where Θ_D is the Debye temperature. The thermal diffuse-scattering intensity (B) is proportional to the temperature T and the Debye-Waller factor as

$$B \propto T \cdot \exp(-2M) \qquad (2)$$

when $T > \Theta_D$. Then, $S/B \propto 1/T$ when $T > \Theta_D$ and S/B=Constant when $T < \Theta_D$ (Θ_D=600K was used.). In the comparison, the S/B values by experiments and by calculation were normalized at RT(300K). Considering those temperature dependent factors and using the intensity at RT as the initial value, the temperature dependence of S/B (●) was calculated.

At 1000ºC (1273K) the ratio decreased to half at RT. The decrease of the S/B ratio is mostly caused by the increase of B that is the thermal diffuse-scattering.

Fig. 4 shows the CTR spectra measured at RT for 100min, at 1000ºC for 100min, and at 1000ºC for 200min. The backgrounds (mostly due to the thermal diffuse-scattering) are subtracted. To increase the S/B ratio at 1000ºC the CTR scattering was measured for 200min. The spectrum quality looks better than that at

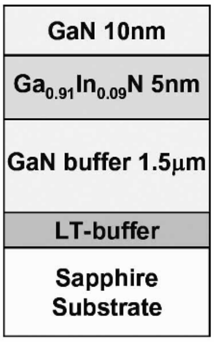

Fig. 2 Sample structure to measure the temperature dependence of S/B ratio.

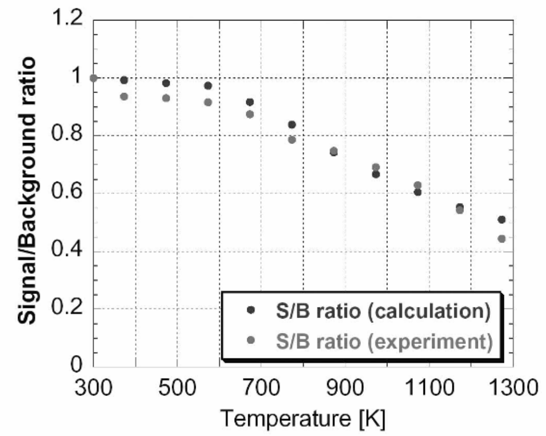

Fig. 3 The temperature dependence of S/B ratio from the sample (●) shown in Fig. 2. (○) is a calculation changing the temperature dependent parameters in the CTR scattering and the thermal diffuse-scattering. The intensities at RT (300K) were used as initial values.

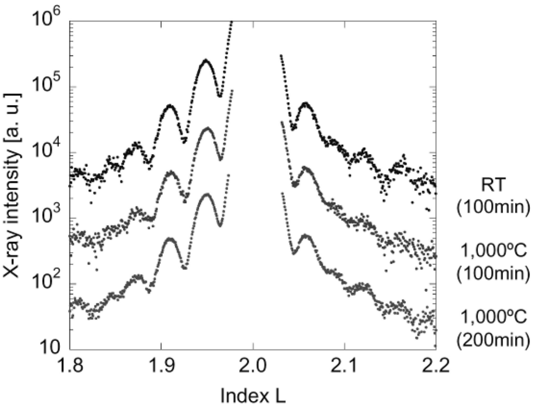

Fig. 4 CTR spectra at RT measured for 100min, at 1000ºC for 100min, and at 1000ºC for 200min. Background (thermal diffuse-scattering) is subtracted.

RT for 100min.

3.2 Spectra at high temperatures and curve-fitting

Two different wafers, as shown in Fig. 5, were used to measure the temperature dependence of the CTR spectra. Measured CTR spectra at several temperatures and at RT before and after the heating are shown in Fig. 6 in the case of GaN/GaInN/GaN on sapphire substrate. Fig. 7 is the similar spectra from GaInN/GaInN/GaInN on GaN substrate. Gray lines in both figures are the best-fit curves.

The fitting results were better in the case of GaInN/GaInN/GaInN on GaN substrate. It is visually clear and also indicated by the R-factor values in both figures. There was curving of the wafer of GaN/GaInN/GaN on sapphire substrate probably due to the difference in the thermal expansion coefficients between GaN and sapphire, even at RT, and should change with temperature. In the calculation those effects were not included (curving should be

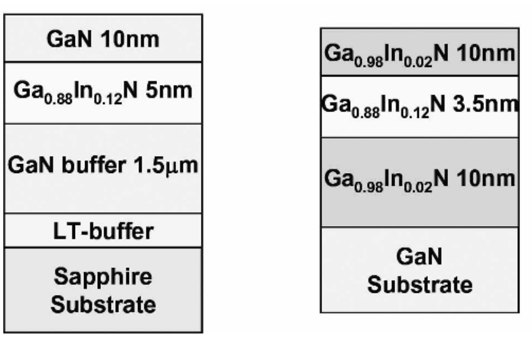

Fig. 5 Two different wafers (left; GaN/GaInN/GaN on sapphire substrate and right; GaInN/GaInN/GaInN on GaN substrate), used for temperature dependence measurements of the CTR spectra.

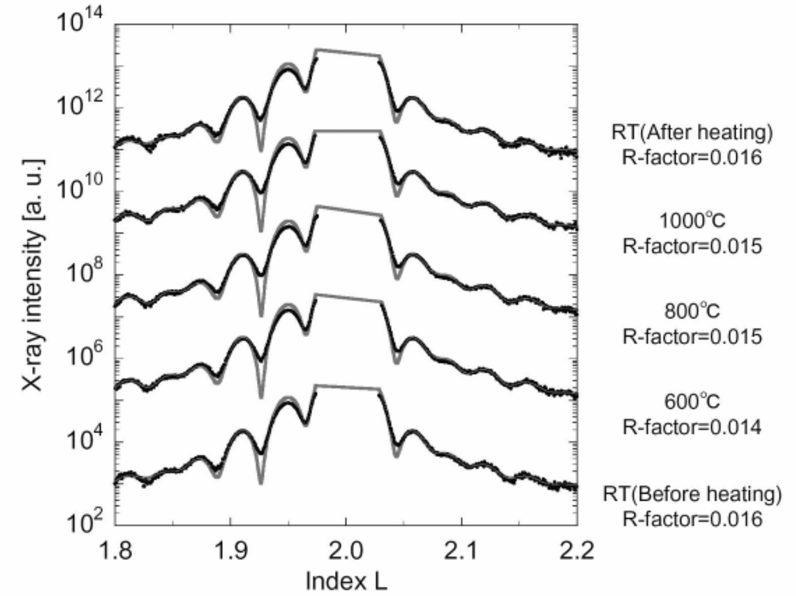

Fig. 6 CTR spectra from GaN/GaInN/GaN on sapphire substrate at several temperatures and at RT before and after heating. Gray curves are best-fit curves to each spectrum. R-factor indicates the fitting quality.

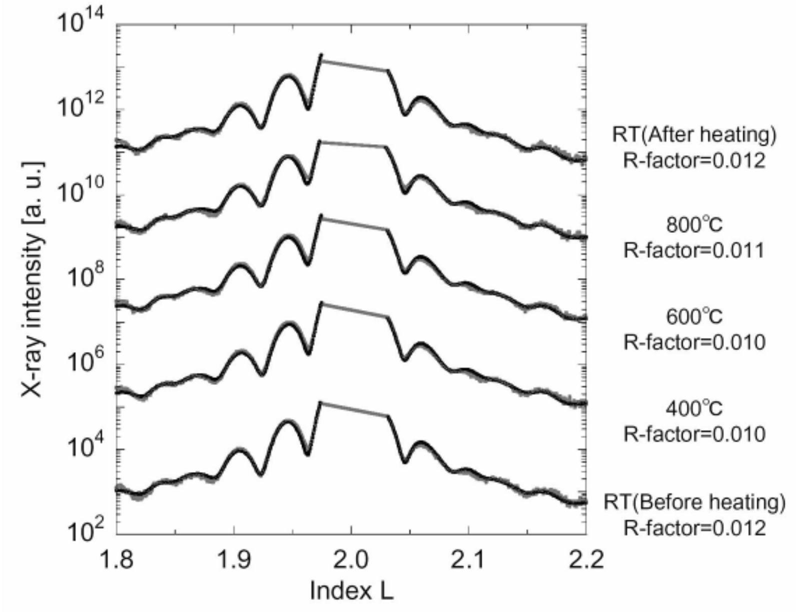

Fig. 7 CTR spectra from GaInN/GaInN/GaInN on GaN substrate at several temperatures and at RT before and after heating. Gray curves are best-fit curves to each spectrum. R-factor indicates the fitting quality.

temperature-dependent since the velocity of the misfit dislocations is also temperature-dependent) and they may affect the fitting quality.

3.3 Composition profiles

Indium (In) composition profiles are shown in Figs. 8 and 9. Fig. 8 shows In composition profiles in GaN/GaInN/GaN on sapphire substrate at RT before heating and at 1000ºC obtained from Fig. 6. Fig. 9 shows In composition profiles in GaInN/GaInN/GaInN on GaN substrate at RT before heating and at 800ºC obtained from Fig. 7.

In both cases, there looks no significant difference between profiles at RT and at high temperatures. In compositions at peak are much lower, in both cases, than those designed (In compositions in Fig. 5 are those values designed.). This difference has been our experience in other many samples. Our understanding is that the In composition is determined from X-ray diffraction, XPS, and/or AES on *thick layers*. However, in a thin (several nm) layers, In is not fully incorporated from gas phase to solid phase. There observed, as in Fig. 7 and 8, a tendency that In increases to the growth direction.

This effect is our first target to investigate using the measurement system described in this paper.

The safety facilities are being designed and installed just in case the Be windows abruptly break at a raised susceptor temperature in a hydrogen and metalorganic source gases.

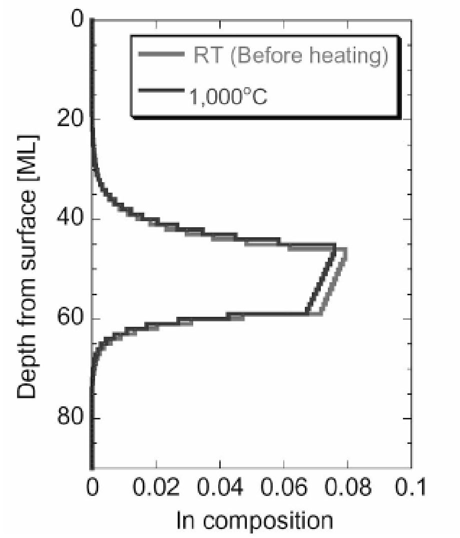

Fig. 8 In composition profiles in GaN/GaInN/GaN on sapphire substrate at RT before heating and at 1000ºC obtained from Fig. 6.

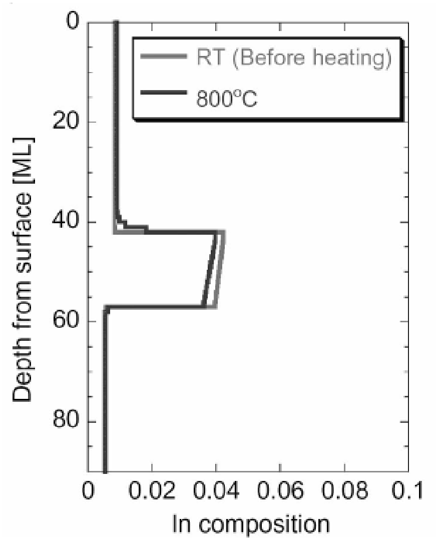

Fig. 9 In composition profiles in GaInN/GaInN/GaInN on GaN substrate at RT before heating and at 800ºC obtained from Fig. 7.

4. SUMMARY

A detailed structure of the reactor and the XYZ-stage installed in the X-ray diffraction measurement system was described, and the proper operation of the whole measurement system on the Ga(In)N/GaInN/Ga(In)N heterostructure at as high as 1000ºC was reported. Though there are several ambiguities in the analysis due to strain in the wafer, very different In profiles were obtained for the case of Ga(In)N/GaInN/Ga(In)N on sapphire and that on GaN.

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