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Analysis of In Distribution in GaInN/GaN Multilayer Structures by X-ray CTR Scattering

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Using X-ray CTR scattering measurement, Ga(In)N/GaInN/Ga(In)N quantum-well structures were investigated and ability of the X-ray CTR scattering measurement on the study of Ga(In)N/GaInN/Ga(In)N multilayer structures was discussed. Though it was difficult to study the GaInN/GaN layers grown on sapphire substrates, the GaInN/GaN layers grown on GaN substrates were successfully investigated by using the X-ray CTR scattering measurement. The results showed that degree of In distribution into the GaN layer was larger when the growth temperature was higher. The In compositions in the cap layers were always twice as large as those in the buffer layers, although they were designed to be the same. It suggests segregation of In from the well layers of which In composition was higher than those of cap and buffer layers.

Key words: GaInN/GaN, quantum-well structure, distribution of In, X-ray CTR scattering

1. INTRODUCTION

GaN-based LEDs can cover blue and green light in the visible region. In the GaN-based blue/green LEDs, GaInN layers play an essential role as active layers. However, it is well known that homogeneous GaInN layers are difficult to grow, since In segregation is often observed in the layers. Therefore, the characteristics of the GaInN layers should be studied more in order to control the layer structures precisely.

We have demonstrated that X-ray CTR (crystal truncation rod) scattering measurement is a very powerful technique to study heteroepitaxially-grown semiconductor layers non-invasively at an atomic scale. The X-ray CTR scattering is a rod-like distribution of scattered X-ray intensity around a Bragg point which is caused by abrupt truncation of the periodicity of a crystal at a surface. Since the structure of over layers grown on the surface modulates the scattered X-ray intensity in the rod-like distribution, information on the structure of the grown layers can be deduced from the X-ray CTR scattering spectra [1-6]. Using the X-ray CTR scattering measurement, layer thicknesses, composition profiles, surface roughness, and deformations of lattice can be analyzed. The X-ray CTR scattering measurement has already been conducted to study the *initial* growth stage of GaInN/GaN/LT(low temperature deposited)-AlN/sapphire structures [7-9].

In this work, we challenge to study Ga(In)N/GaInN/Ga(In)N quantum-well structures using the X-ray CTR scattering measurement, and discuss ability of the X-ray CTR scattering measurement on the study of Ga(In)N/GaInN/Ga(In)N quantum-well structures.

2. EXPERIMENTAL

When a Ga(In)N/GaInN/Ga(In)N structure sample is grown on sapphire substrate as usually conducted with some buffer layers, strong diffuse scattering of X-ray is observed around each Bragg point which is caused by the high-density misfit dislocations and/or

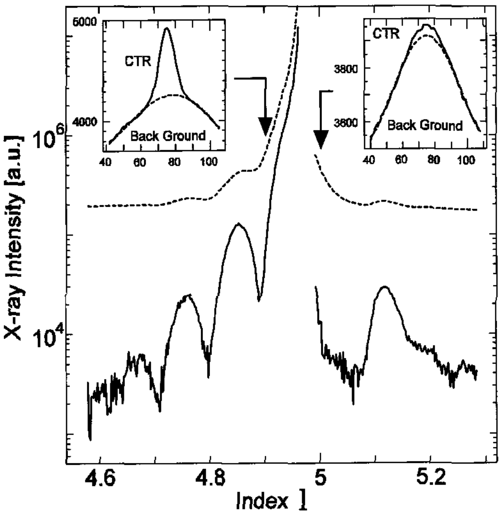

Fig. 1 Measured ω-2θ spectrum (broken line) and CTR scattering(solid line) of GaN/GaInN/GaN structure grown on a sapphire (0001) substrate with LT-deposited AlN buffer layer. l is index defined for sapphire substrate. Insets show rocking curves measured around l=4.9 and 5.0.

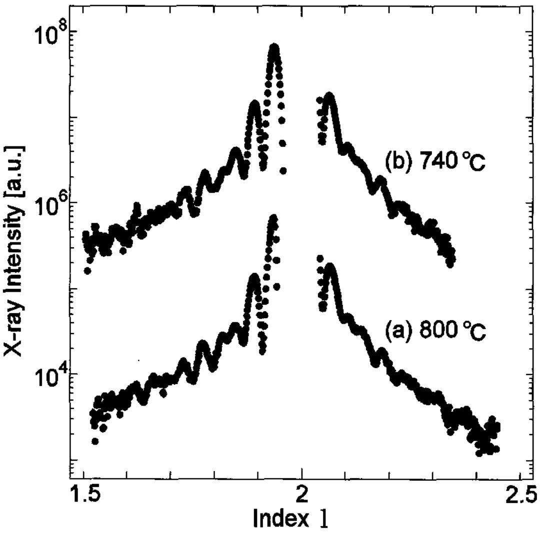

Fig. 2 Measured X-ray CTR scattering spectra of the samples grown on GaN substrates at different growth temperatures, (a) 800 and (b) 740℃. l is index defined for GaN substrate. No significant overlapping of diffuse scattering was observed in the measured region.

amorphous-like or low-crystalline-quality buffer layers. The diffuse scattering, as a back-ground noise, disturbs obtaining a good X-ray CTR scattering spectrum. Figure 1 shows an example of the X-ray CTR scattering spectrum of GaN/GaInN/GaN structure grown on a sapphire substrate. As shown in the insets in Fig. 1, relatively strong diffuse scattering overlapped on the CTR scattering and made it difficult to separate the CTR scattering from back-ground noise. Therefore, the X-ray CTR scattering spectrum shown in Fig. 1 should be suspected.

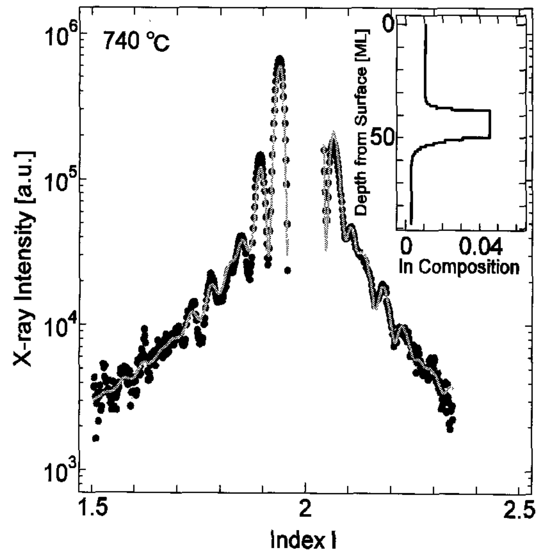

Fig. 3 Measured (black dots) and calculated (gray line) X-ray CTR scattering spectra of the samples grown at 740℃. Inset shows the In distribution which gives the calculated spectra. The In compositions in the well and the barrier layers are roughly equal to the designed values.

$Ga_{0.99}In_{0.01}N$(cap)/$Ga_{0.94}In_{0.06}N$(well)/$Ga_{0.99}In_{0.01}N$ (buffer) structures were prepared by OMVPE. All the samples were grown on GaN substrates in order to avoid the back-ground scattering. Thicknesses of the layers were 10, 3.5, and 10 nm for the cap, well, and buffer layers, respectively. Samples grown at different temperatures, 800, 770 and 740℃, were investigated. The X-ray CTR scattering measurement was conducted at BL6A of Photon Factory in High Energy Accelerator Research Organization at Tsukuba. Wavelength of the X-ray was set at 0.1nm. A Weissenberg camera was used to record the X-ray CTR scattering intensity around the 0002 Bragg diffraction spot of GaN with a CCD camera as a detector.

3. RESULTS AND DISCUSSIONS

As shown in Fig. 2, using the GaN substrate, clear X-ray CTR scattering spectra were obtained. No significant overlapping of back-ground diffuse scattering was observed. The fine structures on the spectra were caused by the interference of X-ray scattered by the multi-layers. The spectra were analyzed by curve fitting method assuming a model structure. Quality of the curve-fitting is estimated using R-factor, defined as

$$\text{R-factor} = \sum_i \frac{|\log(f_i) - \log(g_i)|}{|\log(f_i)|}$$

where f_i and g_i are measured and theoretically-calculated X-ray CTR scattering intensity at a point i. The R-factor for the measured spectra was about 0.02, which indicated that the parameter fitting was conducted well on all the samples.

Figure 3 shows an example (gray line) of the

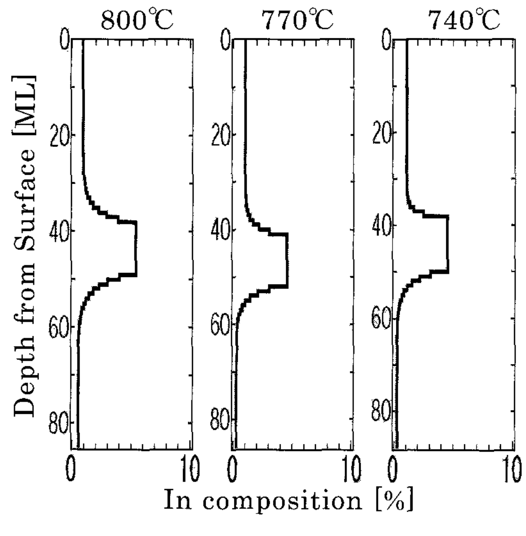

Fig. 4 Distributions of In in the samples grown at 740, 770 and 800℃ analyzed by using the X-ray CTR scattering measurement. It was observed that when the growth temperature was higher, the degree of the distribution of In into the cap layer is greater, as expected.

calculated X-ray CTR scattering spectrum from a model structure with a measured spectrum(dots). Inset shows the In distribution at an atomic scale which gives the calculated spectrum. As shown in Fig. 3, the calculated spectrum was well fitted to the measured one, and as shown in the inset, the measured In compositions in the cap and buffer GaInN layers and well layer were roughly equal to 0.01 and 0.06 as designed. The result suggests that the X-ray CTR scattering measurement is well applicable also for the GaInN/GaN systems even when the In composition is less than 0.1.

Figure 4 shows In distributions in the samples grown at 740, 770, and 800℃. It was found that the degree of In distribution into the GaN cap layer was larger in the sample grown at a higher temperature (800℃) than in the sample grown at a lower temperature (740℃) as expected. The degree of the In distributions are listed in Table I. As listed in Table I, In atoms distributed in the cap layer about 2ML when the growth temperature was high, while it was only 1ML when the growth temperature was low.

These results which agree with our expectation also suggested that the X-ray CTR scattering measurement is a very powerful technique to investigate the GaInN/GaN systems.

Figure 5 shows the In distributions in different scale to precisely observe the In distributions in the cap and the buffer layers. The In compositions in cap layers were larger than those in the buffer layers for all the samples. It might be caused by segregation of In into the cap layers from the well layers. In other words, when a GaInN layer grows on another GaInN layer, the In composition in the upper layer may be higher near the interface than designed composition, and it may gradually decreases to the designed one.

Since the X-ray CTR scattering measurement is a so useful technique to study the GaInN/GaN systems, it is important to develop a new method to measure proper X-ray CTR scattering spectra from the layers grown on sapphire substrates also. One of the possibilities is to measure X-ray CTR scattering spectra around asymmetric Bragg points to make the irradiation angle small and the penetration depth of the X-ray short, not to be suffered from the diffuse scattering from the buffer layers.

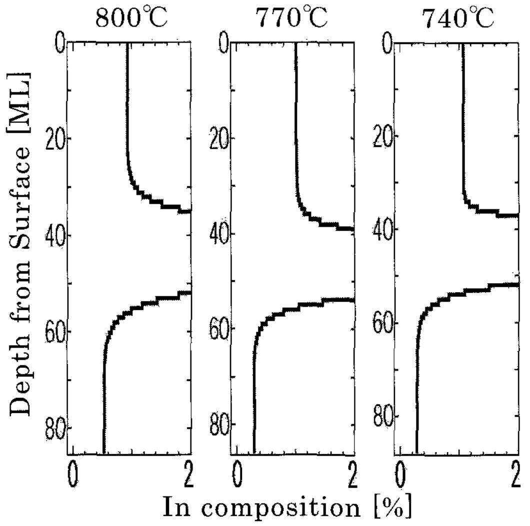

Fig. 5 Distributions of In drawn in larger scale to observe the difference of the In distributions between in the cap and the buffer layers. The In compositions in the buffer layers were as designed, although the In compositions in the cap layers were twice as large as those in the buffer layers.

Table I In compositions in the cap, well and buffer layers, and degree of In distribution into cap and buffer layers from well layer obtained by analyzing the X-ray CTR scattering spectra. The In compositions so low as 1% was well measured. It was shown that the degree of In distribution was large when the growth temperature was high.

Growth Temp. [℃]	In Composition [%]			Distribution of In [ML]	
	Cap	Well	Buf.	Cap	Buf.
800	0.9	5.4	0.5	2.4	2.9
770	1.0	4.5	0.3	1.8	2.3
740	1.1	4.8	0.3	1.1	2.4

4. CONCLUSIONS

Ga(In)N/GaInN/Ga(In)N quantum-well structures were investigated using X-ray CTR scattering measurement, and ability of the X-ray CTR scattering measurement on the study of Ga(In)N/GaInN/Ga(In)N quantum-well structures was discussed. Though it was difficult to study the GaInN/GaN layers grown on sapphire substrates, the GaInN/GaN layers grown on GaN successfully investigated by the X-ray CTR scattering measurement. Since the X-ray CTR scattering measurement was shown to be useful to study the GaInN/GaN structures, it is desired to develop a new method to analyze the layers grown on sapphire substrates also. The results showed that degree of In distribution into the GaN layer was larger when the growth temperature was higher. The In compositions in the cap layers were always twice as large as those in buffer layers, although they were designed to be the same. It suggested segregation of In from the well layers of which In composition was higher than those of cap and buffer layers.

5. ACKNOWLEDGEMENTS

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