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Structural and Optical Properties of AlGaInN Grown by MOVPE

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1. Introduction

Quaternary AlGaInN is a promising material for optical device applications in the ultra-violet spectral region [1, 2]. However, in wurzite AlGaInN, the existence of an unstable mixing region has been predicted [3]. In this paper, we investigate the relationship between the structural and the optical properties of AlGaInN and find that a phase separation and a partial relaxation occur with high Al and In content and they deteriorate the optical properties in the shorter wavelength region.

2. Experimental

AlGaInN quaternary epilayers were grown on 1-μm GaN templates (grown at 1010°C) formed by a standard two-step nucleation procedure with low-temperature GaN grown at 550°C on c-plane sapphire substrates by low-pressure (300 Torr) metalorganic vapor phase epitaxy (MOVPE). The growth temperature of the AlGaInN was fixed at 845°C. Trimethylgallium (TMGa), trimethylaluminium (TMAl), and trimethylindium (TMIn) were the group III sources and NH_3 was the group V source. The thickness of the AlGaInN epilayers was 0.2-0.35 μm. $Al_yGa_{1-x-y}In_xN$ layers with different quaternary compositions (0<y<0.66, 0<x<0.08) were grown by varying TMAl flow rate while keeping the TMGa and TMIn flow rates constant. Compositions of In and Al in the films were measured using Rutherford backscattering spectrometry (RBS) with a 2.275 MeV He^{++} ion beam. Room temperature (RT) photoluminescence (PL) was measured using an excimer (ArF) laser (λ=193 nm) as the excitation source. A high-resolution X-ray diffractometer (Philips X'Pert System) was used to evaluate the structural quality and determine the in-plane and out-of-plane lattice parameters. For the latter, symmetric (00.2) diffraction and reciprocal space map (RSM) on an asymmetric (10.5) reflection were used.

3. Results and Discussion

Four $Al_yGa_{1-x-y}In_xN$ samples were grown under various TMAl flow rates. By increasing TMAl flow rate and keeping the TMIn flow constant, In (x) content in the layers increased from 0.05 to 0.08 with increasing Al (y) from 0.26 to 0.66. This is due to the enhancement of In incorporation efficiency under higher TMAl flow. Figure 1 shows RT PL spectra for AlGaInN layers. The PL peak energy in $Al_{0.26}Ga_{0.69}In_{0.05}N$ [sample (a)] was 3.584 eV, which was fairly close to the theoretical data in J. Han et al. [2]. The increase of Al and In content in samples (b) and (c) resulted in a blue shift of approximately 150 meV. However, no blue shift was observed when the Al and In [sample (d)] were further increased. The emission intensity decreased monotonically with increasing Al and In content. Hirayama et al. reported a decrease of intensity below 340 nm of emission wavelength in 77-K PL spectra of AlGaInN [1].

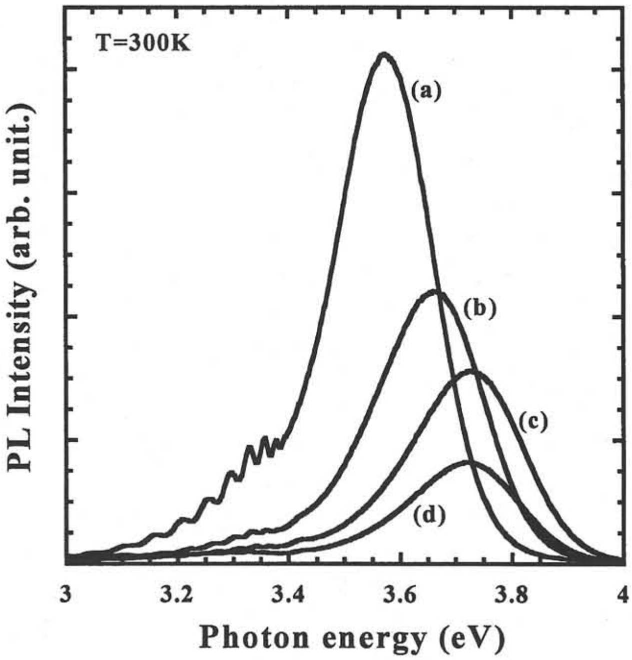

Fig. 1 Room temperature PL spectra of AlGaInN layers. Sample (a) : $Al_{0.26}Ga_{0.69}In_{0.05}N$, (b) : $Al_{0.39}Ga_{0.55}In_{0.06}N$, (c) : $Al_{0.52}Ga_{0.41}In_{0.07}N$, (d) : $Al_{0.66}Ga_{0.26}In_{0.08}N$.

Structural properties of the AlGaInN samples [(a), (b), (c), and (d) in Fig.1] were investigated with a (0002) 2θ-ω X-ray diffraction scan (Fig. 2). Single AlGaInN peak with diffraction fringes was observed for sample (a). The RSM confirmed that the in-plane lattice parameter in sample (a) was the same as that of the underlying GaN layer. These results indicate that the AlGaInN was grown pseudomorphically on GaN layer. With increasing Al and In contents, the AlGaInN diffraction peak [samples (b), (c), and (d)] shifted toward the higher-angle side and the peaks broadened. The degree of peak shift becomes smaller. In addition, another broad peak for lower-angle side was clearly observed in samples (c) and (d). The intensity of the broad peak increased with increasing Al and In contents. In GaInN molecular beam epitaxy growth, a strong phase separated peak has been observed by X-ray diffraction and the phase separation was directly confirmed by a transmission electron microscopy analysis [4]. Therefore, we believe that the other lower-angle broad peak is caused by the phase separation in AlGaInN. In sample (d), RSM measurement showed a in-plane shift of the AlGaInN with respect to the GaN layer, which corresponds to a partial relaxation. From these results, it is clear that, at higher Al (y>0.52) and In (x>0.07) contents, phase separation and partial relaxation in the AlGaInN layer occur and they deteriorate the optical properties.

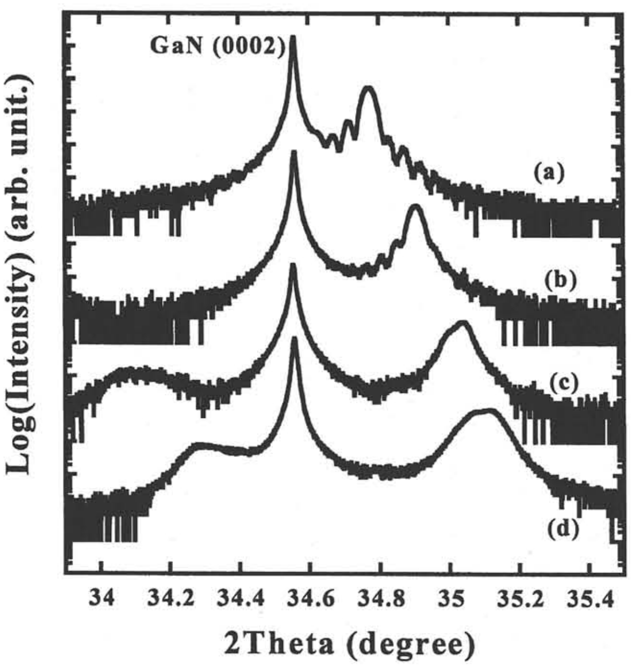

Fig. 2 (0002) 2θ-ω X-ray diffraction of AlGaInN epilayers.

Figure 3 shows in-plane lattice parameter mismatch dependence of the critical layer thickness of AlGaInN layers on GaN. The thickness for AlGaN is also plotted as a reference [5]. The AlGaN layers were grown on a GaN layer at 1010°C. The horizontal axis is defined as $(a_{GaN}-a_{AlGaInN})/a_{GaN}$. The a_{GaN} is the in-plane lattice parameter in GaN (a_{GaN}=0.3188 nm). The $a_{AlGaInN}$ is the relaxed in-plane lattice parameter of AGaInN predicted by Vegard's law. The critical thickness was determined by RSM measurement. In AlGaN growth, the critical thickness decreases sharply with increasing mismatch of in-plane lattice parameters. In contrast, it becomes large in AlGaInN compared with AlGaN. In addition, no crack was observed for AlGaInN samples, even for the relaxed layers. A possible reason for these is the relatively low growth temperature (845°C) or an effect of Indium incorporation.

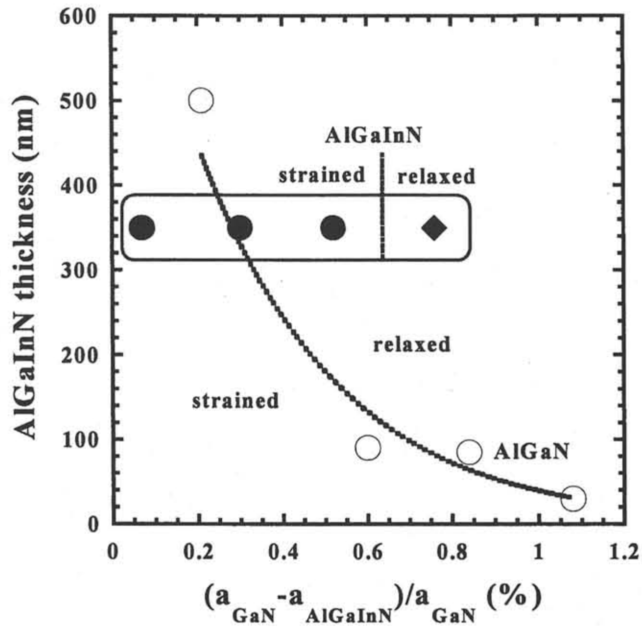

Fig. 3 The in-plane lattice parameter mismatch dependence of critical layer thickness of AlGaInN.

4. Conclusions

The structural and optical properties of AlGaInN layers grown by MOVPE were investigated by RT PL and X-ray diffraction. Phase separation and partial relaxation occurred in AlGaInN with high Al and In content and they deteriorated the optical properties.

Acknowledgments

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