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Phase separation of MOVPE-grown thick (~1 μm) $In_xGa_{1-x}N$ (x~0.3) (I)
——— Simultaneous emergence of metallic In-Ga and GaN-rich InGaN ———

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Introduction: According to the phase diagram of InGaN proposed by Ho and Stringfellow [1], spinodal decomposition should result in the simultaneous formation of InN-rich InGaN and GaN-rich InGaN. However, Doppalapudi et al. [2] did not find such simultaneous formation of the two phases. They found only InN-rich $In_xGa_{1-x}N$ (x~0.97) in as-grown $In_{0.35}Ga_{0.65}N$ film, while only GaN-rich InGaN (2θ~34.5º) was observed after the annealing of the film at 675ºC. Thus, phase separation of InGaN has not yet to been completely understood, in spite of many works. In this work, we report the simultaneous formation of metallic In-Ga and GaN-rich InGaN as a result of phase separation of MOVPE $In_xGa_{1-x}N$ at 650 ºC.

Experiments: The growth of $In_xGa_{1-x}N$ alloys (x~0.3) was conducted using a MOVPE system. TEG, TMI, and NH_3 were used for Ga, In, and N sources, respectively. Growth pressure was fixed at 150 Torr. Growth rate of InGaN was about 0.7 μm/h.The growth process is shown Fig.1 (a). After the annealing of AlN/Si substrate wafers at 950ºC in NH_3, an $In_xGa_{1-x}N$(x=0.34) layer was deposited at 650ºC for 60 min (1st InGaN growth) on three wafers. After cooling down the wafers to RT, Sample A was taken out. Then, the second $In_xGa_{1-x}N$ growth was performed for 30 min at 650ºC. Again, one of the two wafers (Sample B) was taken out. Finally, the remaining wafer was annealed at 650ºC in $NH_3$for 30 min (Sample C).

Results and discussion: Fig.1 (b) shows XRD 2θ/ω profiles for the Samples A, B and C. Note that InN content in the epitaxial $In_xGa_{1-x}N$ was decreased from 0.34 to 0.32 when growth time was increased from 60 to 90 min. When film thickness was increased to 1.1μm (90 min for Sample B) under the same growth conditions, new three peaks appeared as shown in Fig. 1(b), indicating the occurrence of phase separation. These new peaks at 2θ~31.5º, 2θ~32.9º and 2θ~34.5º are very close to those for InN (0002), metallic In (101) and GaN (0002), respectively. These peaks at 2θ~31.5ºand 2θ~34.5º are not due to pure InN and GaN, respectively. Therefore, they are denoted as InN-rich $In_{1-y}Ga_yN$ (0002) and GaN-rich $In_zGa_{1-z}N$ (0002). By the precise measurement of lattice constant by XRD, y and z were determined to be 0.02-0.03 and 0.02-0.05, respectively. The InN composition~0.03 in the GaN-rich InGaN is in good agreement with the solubility of InN in GaN at 650ºC predicted by Ho and Stringfellow [1]. The results in Fig. 1(b) show that the epitaxial $In_xGa_{1-x}N$ (x~0.3) is consumed to form the Ga-rich $In_{1-y}Ga_yN$ and metallic

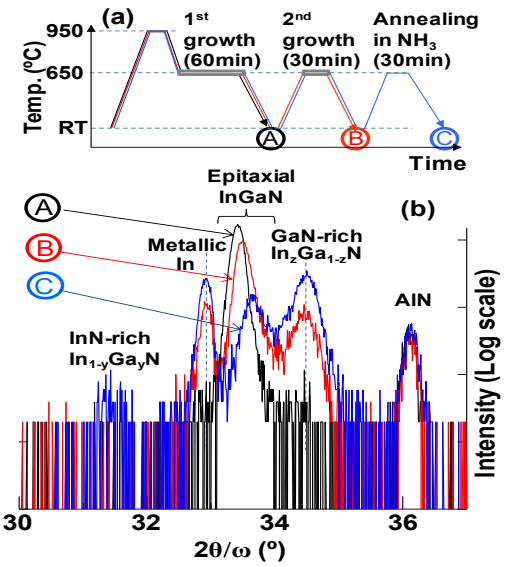

Fig. 1.Time chart of the preparation for three different samples, A, B, and C (a), and XRD 2θ/ω profiles for the three samples (b).

In. As seen Fig. 1 (b), InN-rich $In_{1-y}Ga_yN$ (0002) peak appeared occasionally, not always. This seems that the InN-rich $In_zGa_{1-z}N$ is not stable. Therefore, it is speculated that the InN-rich $In_{1-y}Ga_yN$ is changed swiftly into metallic $In_{1-y}Ga_y$. From the EDS measurements, it was shown that the metallic In contained about 3 at% of Ga. As can be seen in , the sample annealed in NH_3 flow at 650ºC (Sample C) is found to markedly enhance the decomposition reaction.

Thus, by using AlN/Si(111) substrates, we have clearly observed the phase separation of epitaxial $In_xGa_{1-x}N$ into metallic In-Ga and GaN-rich $In_zGa_{1-z}N$. It should be pointed out that the selection of substrate and /or interlayer for InGaN growth is critical issue in the observation of such phase separation.

Conclusion: we have clearly observed the simultaneous formation of metallic In-Ga and Ga-rich InGaN during the MOVPE growth of $In_xGa_{1-x}N$ (x~0.3) at 650 ºC, as a result of phase separation. Metallic In-Ga is formed through the In-rich InGaN due to the thermal instability of In-rich InGaN.

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