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Arsenic Surfactant and Incorporation Effects on Cubic GaN Grown by Metalorganic Vapor Phase Epitaxy

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1. Introduction

The III-V-N type alloys are peculiar materials because of their huge bandgap bowing. In comparison to InGaN, which is presently in use as the active layer material for blue laser diodes, N-rich GaNAs alloy is expected to realize a wider range of bandgaps with a smaller lattice mismatch to GaN. Very recently, the molecular beam epitaxy (MBE) growth and photoluminescence (PL) properties of N-rich hexagonal GaNAs (h-GaNAs) alloys were reported and their huge bandgap bowing was confirmed [1]. On the other hand, there have been no reports on the growth of N-rich cubic GaNAs (c-GaNAs) alloys, where the binary components are the same crystal structure. It has been reported that As ambient pressure acts as surfactant in MBE growth [2]. A first-principles total-energy calculation has shown that a submonolayer As coverage on GaN (001) surface reduces the surface energy to stabilize the (001) surface [3]. In order to grow c-GaNAs alloys it is important to clarify the As effects on c-GaN growth, which in metalorganic vapor phase epitaxy (MOVPE) have not been clarified yet.

In this paper, the As effects on c-GaN growth in MOVPE are reported. The As effects on crystal structures were examined by scanning electron microscopy (SEM) and X-ray diffraction (XRD) measurements. The As incorporation in c-GaN epilayers was confirmed by secondary ion mass spectrometry (SIMS). The optical properties of the As-incorporated c-GaN were studied by photoluminescence (PL) spectroscopy.

2. Experimental

The As-incorporated c-GaN samples were grown by low pressure (160 Torr) MOVPE on GaAs (001) substrates. Trimethylgallium (TMG), 1,1-dimetylhydrazine (DMHy) $[(CH_3)_2N_2H_2]$ and arsine (AsH_3) were used as the precursors of Ga, N and As, respectively. A thin GaN buffer layer (20 nm) was first grown on the substrate at 580 °C, followed by the growth of GaNAs layer (AsH_3 flow rate: 0~450 μmol/min) at 920 °C.

3. Results and discussion

When AsH_3 flow rates were less than 225 μmol/min, the surface morphologies of samples were mirror-like. From the plan view SEM observations of the mirror-like samples, it was found that the samples grown in the presence of an AsH_3 flow have smaller and more isotropic grains than c-GaN (without an AsH_3 flow). This suggests that As adsorption on GaN (001) surface lowers the surface energy.

To examine the crystal structure X-ray reciprocal space maps were measured. Figure 1 shows the three dimensional graphs of X-ray reciprocal space maps of (a) c-GaN and (b) c-GaNAs (AsH_3 = 90 μmol/min) with X-ray beams incident along <1$\bar{1}$0> azimuth. The peak from {1$\bar{1}$01} hexagonal GaN (h-GaN) become weaker with AsH_3 flux.

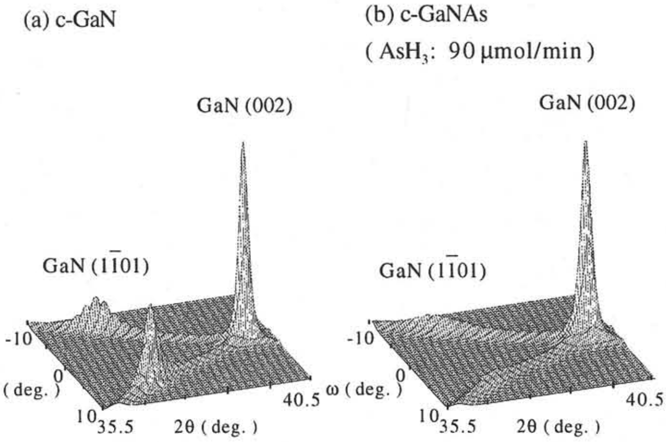

Fig. 1 The three dimensional graphs of reciprocal space maps of (a) c-GaN and (b) c-GaNAs (AsH_3 flow rate = 90 μmol/min) with X-ray beams incident along <1$\bar{1}$0> azimuth.

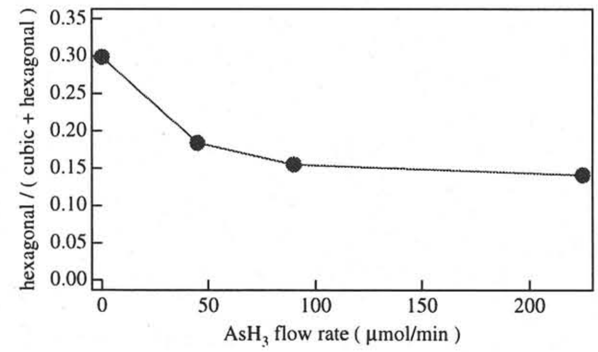

Fig. 2 The inclusion ratio of hexagonal phase in c-GaNAs epilayers against AsH_3 flow rates.

Figure 2 shows the inclusion ratio of hexagonal phase against AsH_3 flow rates estimated by XRD measurements. With increasing AsH_3 flow rates, the inclusion ratio of hexagonal phase decreases. These XRD results clearly show that the As ambience suppresses the inclusion of hexagonal phase in c-GaN epilayers grown by MOVPE.

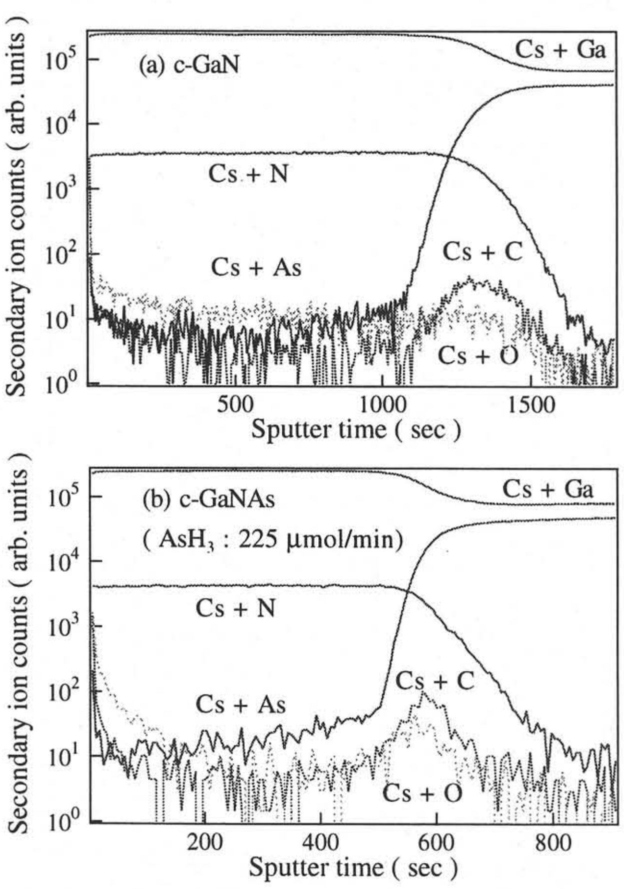

Fig. 3 Secondary ion mass spectrometry (SIMS) profiles of c-GaN and c-GaNAs (AsH_3 flow rate = 225 µmol/min).

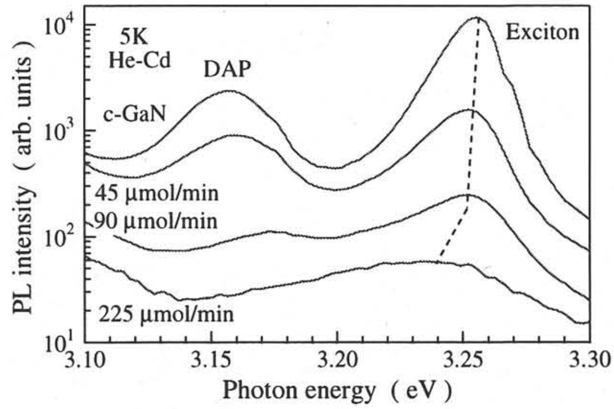

Fig. 4 Photoluminescence (PL) spectra of c-GaNAs grown with various AsH_3 flow rates.

Figure 3 shows SIMS depth profiles of c-GaN and c-GaNAs (AsH_3 flow rate = 225 µmol/min), respectively. Comparing the SIMS profiles, the As incorporation in the samples grown with a AsH_3 flux is confirmed. It was also confirmed that with increasing AsH_3 flow rates, As incorporation increased.

Figure 4 shows the PL spectra of the samples grown with various AsH_3 flow rates. With increasing the AsH_3 flow rate, the exciton peak shifts toward lower energies and become broad, due to As incorporation in c-GaN. The maximum value of the peak shift is 20 meV, which corresponds to a ~0.1% As incorporation as expected from the bowing parameter of h-GaNAs [1].

4. Conclusions

The As effects on the c-GaN growth in MOVPE were studied. It was found that the As ambience significantly suppresses the inclusion of the hexagonal phase in c-GaN. The role of a small amount of As is considered as surfactant. Moreover, a certain amount of As was incorporated in c-GaN with a large AsH_3 flux. This suggests that higher As concentration c-GaNAs alloys can be obtained at growth conditions further from equilibrium, i.e., lower temperatures and/or faster growth rates.

References

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