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Growth of GaNAs films with As_2 source in atomic hydrogen-assisted molecular beam epitaxy

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ABSTRACT

We have investigated the effect of using As_2 source instead of a more commonly used As_4 source on the crystalline quality of GaN_xAs_{1-x} thin films grown by atomic hydrogen-assisted molecular beam epitaxy. Improved structural and optical properties of GaNAs thin films were obtained by using As_2 source. The nitrogen atoms were incorporated into GaAs at a more stable rate under As_2 flux than As_4 flux, and a two-dimensional nucleation growth mode was promoted for growth of GaN_xAs_{1-x} with As_2 source. As a consequence, the surface roughness measured for a 500 nm-thick $Ga_{0.008}N_{0.992}As$ sample grown with As_2 flux was 1–2 monolayers, which was three times more smoother than that for As_4 sample. The photoluminescense measurements showed an improved potential fluctuation of 78.1 meV and twice the intensity at room temperature for As_2 sample.

1. Introduction

Recently, dilute nitride III–V semiconductor alloys are investigated for wide applications ranging from long-wavelength optical communication lasers [1,2] to high-efficiency multi-junction tandem solar cells [3]. The major advantage of this alloy is that the addition of a few atomic percent of nitrogen in GaAs leads to a gigantic reduction in the band gap energy, and can still be lattice-matched to GaAs. However, the inherent metastability of this material leads to generation of high defect densities, compositional inhomogeneity, and rougher interfaces. Consequently, short minority carrier lifetimes [4] as well as radiative emission dominated by localized excitons [5] have been reported. Furthermore, both optical and electrical properties of GaInNAs layers become increasingly degraded with increasing N composition. In order to improve the crystalline quality, there have been reports in the literature to optimize the morphological, structural, and electronic properties of ternary GaNAs as well as quaternary GaInNAs quantum wells (QWs). This has led to 1.3 and 1.5 μm laser diodes on GaAs substrates with excellent device characteristics [6–9]. Furthermore, use of Sb [10] and atomic hydrogen [11] as surfactants in MBE growth have been reported. On the other, it is well known that difference in the sticking coefficients between the arsenic species, As_2 and As_4, affects the adatom migration in the growth of GaAs [12]. However, the difference in the growth mechanism of GaInNAs thin films under different arsenic species has not been fully understood at present. In this work, we studied the effect of using As_2 source instead of a more commonly used As_4 source on the material quality of GaInNAs films grown by atomic H-assisted MBE (H-MBE).

2. Experiments

The GaNAs films were grown on GaAs (0 0 1) substrates by H-MBE using RF-plasma as a nitrogen source as reported elsewhere [11]. After the oxide removal and growth of a 250 nm-thick GaAs buffer layer at 580 °C, a 500 nm-thick GaN_xAs_{1-x} film was grown at 480 °C at a growth rate of 1.0 μm/h. The RF power and nitrogen back pressure was set to 50 W and 3.0×10^{-6} Torr for all growths, respectively. The GaNAs layers were grown either with As_2 or As_4 in order to compare their material qualities. The beam flux of As_2 and As_4 measured at the substrate position was varied from 1.2×10^{-5} to 3.0×10^{-5} Torr. Atomic H was irradiated at a back pressure of 4.0×10^{-6} Torr throughout the growth. The N

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compositions in GaNAs films were determined by using high-resolution X-ray diffraction (HR-XRD), in which the relative shifts of GaNAs Bragg peaks with respect to GaAs peak are determined. The growth process and surface morphology were studied by in situ reflection high-energy electron diffraction (RHEED) and ex situ atomic force microscopy (AFM). The optical properties were investigated by using photoluminescense (PL) measurements, in which PL spectra were recorded in the range between 30 and 300 K using the 532 nm line of a second harmonic generation (SHG) Nd:YVO_4 laser as an excitation source. The PL signals were detected by using a liquid nitrogen-cooled InGaAs photodiode and a standard lock-in technique.

3. Results and discussion

Fig. 1 shows the dependence of N compositions in GaNAs films measured by HR-XRD grown with As_2 or As_4 source as a function of arsenic beam pressure. The average N composition of ~0.85% was obtained under the given arsenic pressure for both As_2 and As_4 sources. However, we note that N composition in samples grown with As_4 source is more sensitive to the arsenic pressure than for As_2. In fact, N composition changes, using As_4 source, from 0.4% at 1.6×10^{-5} Torr to 0.85% at 2.3×10^{-5} Torr, respectively. This suggests that N atoms are incorporated into GaAs at a more stable rate under As_2 source than under As_4 flux.

Next, we evaluated the surface morphology of each sample by using RHEED and AFM. Fig. 2 shows the RHEED patterns along [1 1 0] azimuth after the growth of 500 nm-thick films with (a) As_2 and (b) As_4 source under a beam pressure of 2.0×10^{-5} Torr, respectively. We observe a clear (2×2) RHEED streak pattern for sample grown with As_2, while a weak streak or spotty pattern is observed for sample grown with As_4. Fig. 2(c) shows the RHEED oscillations measured for each arsenic source. A clear RHEED oscillation with near constant amplitude is observed for growth with As_2 flux, while the intensity of RHEED oscillation drops fast as deposition proceeds for As_4 sample. Therefore, the results suggest that a 2-dimensional (2D) nucleation growth mode is promoted by using As_2 source. Fig. 3 shows the surface AFM images and cross-sectional profiles of GaNAs films grown at an arsenic beam pressure of 2.0×10^{-5} Torr for (a) As_2 and (b) As_4 source, respectively. An atomically flat surface and small 2D islands with high step density can be observed for sample grown with As_2. Fig. 4 plots the root-mean-square (RMS) values of surface roughness measured for GaNAs films as a function of As_2 or As_4 pressure as determined by AFM. The RMS value of ~0.4 nm (less than 2 monolayers (MLs)) obtained for sample grown with As_2 is smaller by a factor of 2–4 compared to that of sample grown with As_4 under the same arsenic beam pressure.

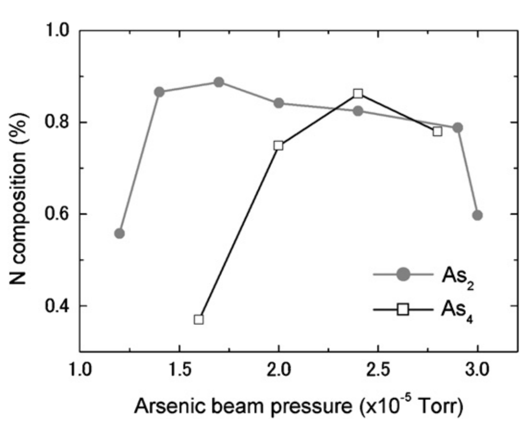

Fig. 1. Dependence of N composition in 500 nm-thick GaNAs films grown with As_2 or As_4 source as a function of arsenic pressure measured by XRD.

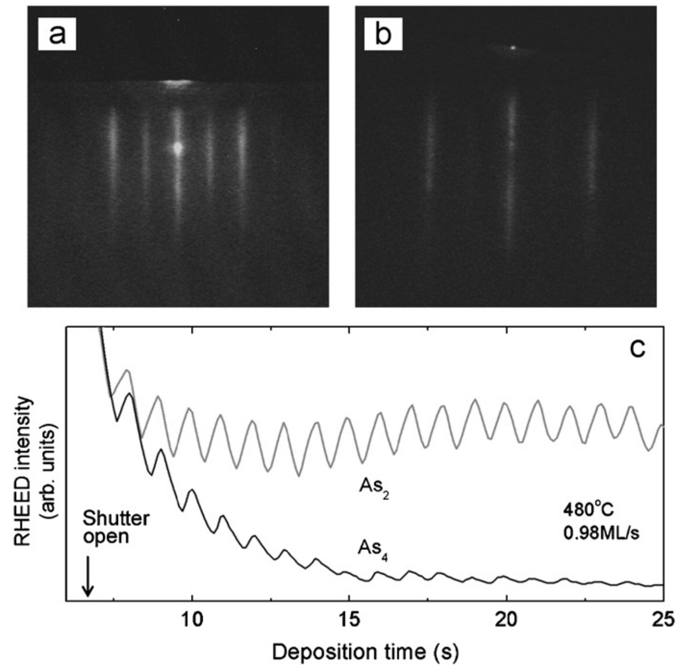

Fig. 2. RHEED images after the growth of 500 nm-thick GaNAs films grown with (a) As_2 and (b) As_4 at a beam pressure of 2.0×10^{-5} Torr, respectively. (c) Shows the RHEED oscillations measured under each As source.

A reduction of surface diffusion of Ga atoms under As_2 source has been reported for growth of GaAs on GaAs (0 0 1) substrates by Sugaya et al. [12]. Since As_2 is a more active species than As_4, we think that an improved surface morphology and reduced alloy disorder in GaNAs films can be obtained with As_2 source compared to As_4. We thus believe that use of As_2 source promotes 2D nucleation growth mode and incorporation of N atoms into the steps of small 2D islands is facilitated thereby resulting in an atomically smooth surface. One further point to mention is that the surface morphology degrades at low As_2 pressures as seen in Fig. 4 and this is due to an enhanced alloy disorder and/or phase separation caused by an increase of surface migration under a low arsenic flux.

Next, we investigated the temperature dependence of PL peak energy in order to evaluate the degree of compositional inhomogeneity. The PL peak energy as a function of temperature for each sample is plotted in Fig. 5. The excitation intensity was ~1 W/cm^2. In general, dilute nitride films typically show anomalous temperature dependence [5]. In the presence of potential fluctuations, the excitons can become trapped in the deepest potential fluctuation at low temperatures. As the temperature is raised, some may escape out and recombine at delocalized states leading to high band-to-band recombination efficiency. This leads to a commonly observed S-shape dependence [5]. In our samples, the PL emission from localized states around 1.20 eV is observed below 120 K and emission from delocalized states becomes dominant at higher temperatures. Here, we define ΔE as a degree of potential fluctuation, in which ΔE is the amount of emission energy shift from the localization to a full delocalization state. ΔE for the sample grown with As_2 source improved to 78.1 meV from 90.9 meV for sample grown with As_4 source. This indicates that use of As_2 source is effective in reducing the potential fluctuation and compositional inhomogeneity.

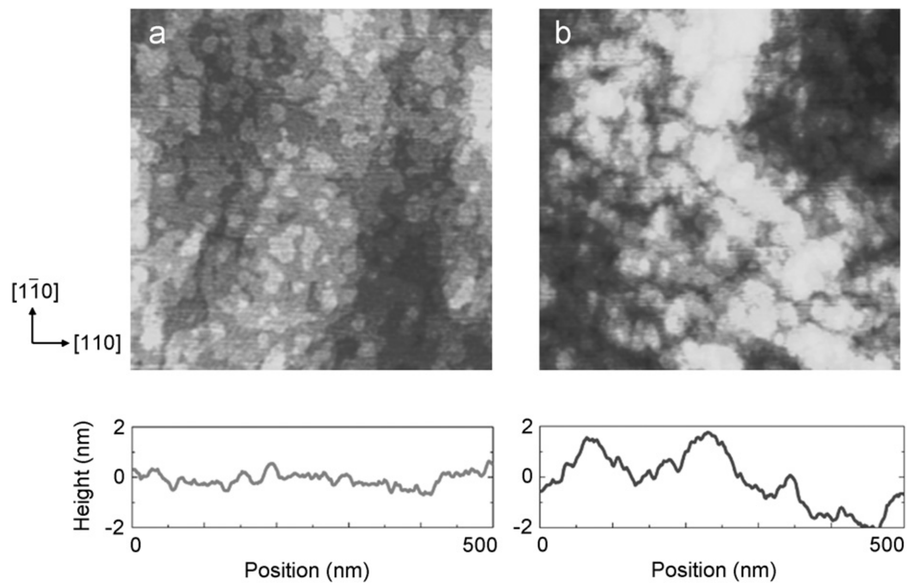

Fig. 3. AFM images and cross-sectional profiles of the GaNAs films grown with (a) As_2 and (b) As_4, respectively. Scan size is 500×500 nm.

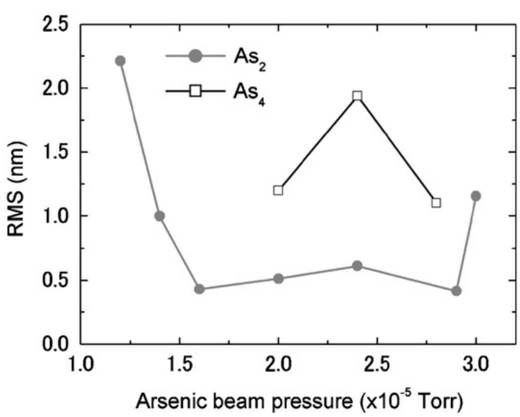

Fig. 4. Dependence of surface roughness (RMS) for 500 nm-thick GaNAs films grown with As_2 and As_4 source, respectively, as a function of arsenic pressure.

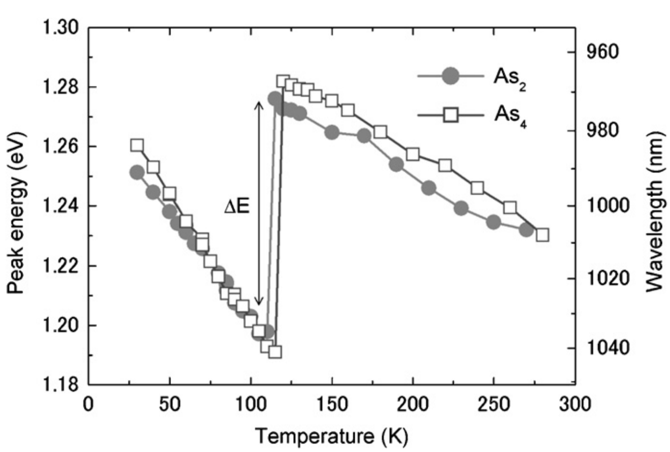

Fig. 5. PL peak energy as a function of temperature measured for GaNAs films grown with (a) As_2 and (b) As_4, respectively, at a beam pressure of 2.0×10^{-5} Torr.

Finally, Fig. 6 shows the PL spectra measured at RT for samples grown with each arsenic source. Though PL emission from the delocalized states, or band-to-band recombination, are clearly observed around 1.26 eV, the integral PL intensity for As_2 sample is twice as large as that for As_4. This suggests that the density of defects and non-radiative recombination centers are reduced for samples grown with As_2 source.

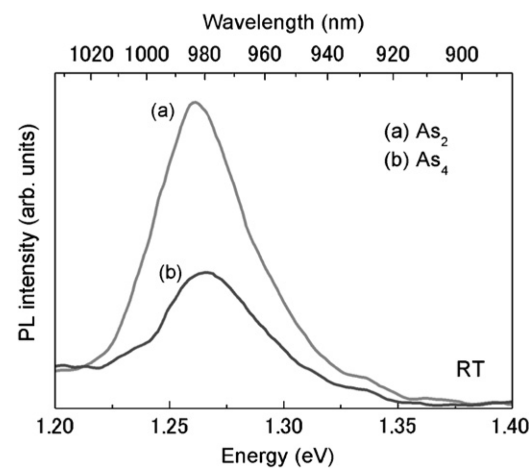

Fig. 6. PL spectra at RT for the samples grown with each As source under the same beam pressure of 2.0×10^{-5} Torr.

4. Conclusion

We have investigated the difference in the structural and optical properties of $Ga_{0.008}N_{0.992}As$ thin films grown by As_2 and As_4 in H-MBE. We were able to fabricate a higher crystalline quality by using an As_2 source instead of more conventionally used As_4 source. By using As_2 source, we find that 2D nucleation growth mode is promoted, and N atoms are incorporated into the steps of 2D islands more favorably resulting in smoother surface. The surface roughness for a 500 nm-thick GaNAs of 0.416 nm, and the potential fluctuation of 78.1 meV has been obtained. Further, the PL intensity at room temperature was twice as strong compared to sample grown with As_4, which thus indicates that non-radiative recombination is reduced. Though the findings described here might be valid just for the given growth condition, further experiments on the effect of arsenic species on GaInNAs growth are currently undertaken.

Acknowledgments

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