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Growth Temperature Dependence of Nitrogen Incorporation in GaNAs Grown by Chemical Beam Epitaxy

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1. Introduction

Recently, considerable attention has been paid to GaNAs and InGaNAs for long wavelength applications. Lasers based upon InGaNAs/GaAs with the nitrogen content of about 1%, operating at a wavelength of 1.3 µm, have been demonstrated. Many authors have tried to find out the growth mechanism of nitrogen incorporation. Researchers have observed up to two temperature dependent regions for nitrogen incorporation, depending on growth methods and precursors used. Auvray *et al.*[1)] studied the effects of growth temperature on the nitrogen incorporation in GaN_xAs_{1-x} grown by MOCVD using TMG, AsH_3, and DMHy as precursors and found two obvious temperature dependent regions for nitrogen incorporation with activation energies of 3.7 and 0.6 eV, respectively. The observed transition between the two regions is attributed to an increased desorption of nitrogen at high temperatures.[2)] Moto *et al.*[3)] found only one temperature dependent region for nitrogen incorporation in GaN_xAs_{1-x} grown by MOCVD. Jin *et al.*[4)] found that there is only one temperature dependent region for nitrogen incorporation in GaN_xAs_{1-x} grown by CBE using TEG, DMHy and solid arsenic (As_4) as precursors. The activation energy is calculated to be 0.97 eV.

In this paper we investigate the effects of growth temperature on the nitrogen incorporation in GaN_xAs_{1-x} grown by CBE using AsH_3, TEG, and active nitrogen species generated by a radio-frequency coupled plasma source (200W) from N_2 as sources. It is found that there are three obvious temperature dependent regions for nitrogen incorporation in GaN_xAs_{1-x}. The activation energies are calculated to be 0.95, 0.05, and 0.59 eV, respectively, and the incorporation mechanisms at different temperature regions are proposed.

2. Experimental

GaN_xAs_{1-x} epilayers are grown on semi-insulating (001) GaAs substrates. The growth temperatures have been calibrated by a pyrometer. Nitrogen content is determined from XRD assuming the validity of Vegard's law. For all the samples used in the present study, clear fringes can be observed in XRD spectra, showing good crystalline quality. The thickness, and therefore the growth rate (r_g, nm/h) can be calculated. Surface morphologies of the epilayers are observed by AFM.

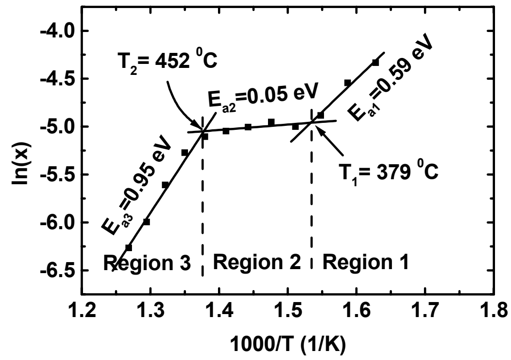

Fig. 1. Dependence of nitrogen composition on growth temperature.

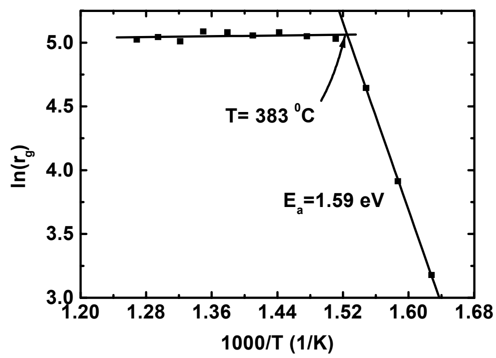

Fig. 2. Dependence of growth rate on growth temperature.

3. Results and discussions

Fig. 1 shows the dependence of nitrogen content in GaN_xAs_{1-x} on growth temperature. The flow rates of TEG, N_2, and AsH_3 are 0.04, 1.25, and 0.75 sccm, respectively. Generally speaking, with increasing temperature, the nitrogen content decreases. However, it can be seen from this figure that three distinct temperature dependent regions can be identified as the growth temperature is increased from 340 to 515°C and the transformation temperatures are calculated to be 379 and 452°C (see Fig. 1), respectively. In addition, at low (Region 1) and high (Region 3) growth temperatures, the nitrogen content decreases very fast (exponentially) with increasing growth temperature, and the activation energies are determined to be 0.59 and 0.95 eV, respectively. At middle temperatures (Region 2) the nitrogen content slightly decreases with increasing growth temperature, and the activation energy is calculated to be 0.05 eV. The growth temperature dependence of nitrogen content in Region 1 is due to TEG pyrolysis controlled process, and the transformation temperature (379°C) from Region 1 to Region 2 is consistent with theoretically estimated pyrolysis temperature of TEG (381°C).[5] This conclusion is also supported by the fact that the transformation temperature (383°C) obtained by the growth temperature dependence of growth rate (see Fig. 2) is in good agreement with the theoretical pyrolysis temperature of TEG. The growth temperature dependence of nitrogen content in Region 2 with the activation energy of 0.05 eV is ascribed to TEG mass transport limited process. This is supported by the fact that the growth rate increases with increasing TEG flow rate and saturates when TEG flow rate is increased to 0.04 sccm (not shown). This conclusion is also supported by the growth rate dependence on growth temperature (see Fig. 2). At high growth temperatures (Region 3), the activation energy 0.95 eV obtained in this work is consistent with 0.97 eV obtained by Jin *et al*.[4] for nitrogen incorporation in GaN_xAs_{1-x} grown by CBE. Because the nitrogen solubility in GaAs is expected to increase with increasing temperature according thermodynamic model, the decrease in nitrogen content with increasing growth temperature is clearly controlled by surface kinetics. The decrease in nitrogen content is mainly determined by the incorporation competition between nitrogen and arsenic in group V sites. One the possible reasons for the decrease of nitrogen content in this temperature range is that with increasing temperature, the pyrolysis efficiency of AsH_3 increases. However, before the introduction of AsH_3 into the growth chamber, the AsH_3 used in our case has been pyrolyzed at 1000°C, so the concentration of the dissociated gases should be constant after using this pyrolysis process according to the experimental results of Calawa.[6] In addition, it can be seen from Fig. 2 that the growth rate is kept constant in this temperature range, strongly suggesting that the residence time of TEG and its fragmented parts are not the limiting process. If it is the limiting process, the growth rate should decrease because of TEG desorption and the nitrogen content should increase. The constant growth rate dependence on growth temperature in Region 3 (see Fig. 2) is also consistent with that of GaAs grown by CBE, suggesting that the growth of GaN_xAs_{1-x} in this temperature range is almost the same as that of GaAs because of low nitrogen incorporation. This result is also supported by AFM observations (not shown because of limited pages). Because arsenic incorporation rate is several hundreds times higher than that of nitrogen, it is suggested that the growth temperature dependence of nitrogen content in Region 3 is due to nitrogen desorption. This conclusion is consistent with the model proposed by C. Jin.[4] Similar results are also found in GaN_xAs_{1-x} grown by MOCVD.[2]

In addition, the above three distinct temperature dependent growth processes are also supported by our AFM results (not shown). Our AFM results clearly show that with increasing growth temperature, there are three types of temperature dependent surface morphologies.

4. Conclusions

In conclusion, three obvious temperature dependent regions for nitrogen incorporation in GaN_xAs_{1-x} grown by CBE are reported for the first time. At low and high growth temperatures, the nitrogen incorporation is TEG pyrolysis controlled and nitrogen desorption controlled process, respectively. The TEG mass transport limited process is found at middle temperatures. The activation energies are determined to be 0.59, 0.95, and 0.05 eV, respectively. The transformation temperatures are calculated to be 379 and 452°C, respectively. Our conclusion is also supported by AFM observations.

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