Original Papers

Repeatability and Mechanisms of Threading Dislocation Reduction in InN Film Grown with In Situ Surface Modification by Radical Beam Irradiation

by

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The objective of this study was to investigate the repeatability of in situ surface modification by radical beam irradiation to reduce threading dislocation density in InN film. The growth of InN template and N radical irradiation processes were repeated twice in situ in the radio-frequency plasma-excited molecular beam epitaxy chamber before the regrowth of InN film on the N radical irradiated template. Transmission electron microscopy was applied to study dislocation behaviors of the InN film grown. In this letter, we show cross-sectional-view transmission electron microscopy evidence of the threading dislocation reduction from \( \sim2.8\times10^{10}\ \text{cm}^{-2} \) in the first irradiated InN layer to \( \sim2.0\times10^{10}\ \text{cm}^{-2} \) in the second irradiated InN layer, and to \( \sim1.3\times10^{10}\ \text{cm}^{-2} \) in the top regrown InN layer. The mechanisms of threading dislocation reduction were also studied.

Key words:

Indium nitride, Molecular beam epitaxy, Threading dislocation

1 Introduction

The interesting properties of InN such as the small bandgap (\( \sim0.65\ \text{eV} \))\(^{1-2} \), the small effective mass (\( \sim0.055m_0 \))\(^{3} \), and the high mobility (\( 14,000\ \text{cm}^2/\text{Vs} \))\(^{4} \) make it suitable for numerous applications including near infrared optoelectronic devices and high-speed electronic devices. When alloyed with GaN or AlN, it ensures light emission covering the whole solar spectrum which is very promising for high-efficiency solar cell application\(^{5} \). In addition, Miller et al. reported a high Seebeck coefficient which is one of the crucial parameters for thermoelectric application in InN\(^{6} \). Up until now, great efforts have been made for the improvement of InN growth\(^{7-11} \). Molecular Beam Epitaxy (MBE) is preferred over Metal Organic Chemical Vapor Deposition (MOCVD) because of the low dissociation temperature of InN\(^{12} \), and so far, the highest-quality InN epitaxial thin films have been obtained by radio-frequency plasma-excited MBE (RF-MBE)\(^{13} \). Our group have invented the droplet elimination by radical beam irradiation (DERI) method for high reproducibility of atomically flat surface InN films\(^{14,15} \). However, the issue of high threading dislocation density in the InN films remains unsolved. It is well known that InN grown on foreign substrates such as sapphire and GaN, contains a high density of threading dislocations in the range of \( 10^{10}\ \text{cm}^{-2} \)\(^{16} \) due to the large difference of the lattice constants between InN and the substrates.

In RF-MBE growth of III-nitrides, the MBE reactor is equipped with an RF plasma generator to supply active N radical as a nitrogen source. Besides as the nitrogen source, several other applications of N radical irradiation have been reported. For example, Gangopadhyay et al. applied the N radical beam irradiation to remove the oxide layer on the surface of GaN film\(^{17} \). They observed a transition from a streaky to a spotty reflection high energy electron diffraction (RHEED) pattern, which indicates a roughening of the GaN surface due to active-nitrogen exposure at a high substrate temperature of 700 °C. Furthermore, Xue et al. made use of in situ annealing and N ion irradiation for nucleation of self-induced InGaN nanocolumn by MBE\(^{18} \). We also recently reported achieving a reduction of threading dislocation density using in situ surface modification by N radical beam irradiation method\(^{19,20} \). In this method, we employed N radical irradiation to modify surface morphology of InN template in situ in the MBE chamber before re-growing InN film on the irradiated template. From transmission electron microscopy (TEM) observation, it was confirmed that some of threading dislocations bent and annihilated at the interface of the irradiated InN layer and the regrown InN film, and thus reduced the threading dislocation density in the top InN film. The repeatability of the proposed method was expected but have not been confirmed experimentally. In this report, we investigated the repeatability of in situ surface modification by radical beam irradiation to reduce threading dislocation density in InN film and we show cross-sectional view TEM evidence of the two stages of threading dislocation reduction. The mechanisms of threading dislocation reduction will also be discussed.

2 Experimental Method

The sample was grown on MOCVD-grown (0001) GaN/sapphire substrate in a conventional RF-MBE system (EpiQuest RC2100NR) equipped with conventional Knudsen
cells of group-III metals and a nitrogen plasma source (SVT Associates 6.03). The growth time chart is as shown in Fig. 1. Prior to growth, the MOCVD-grown (0001) GaN/sapphire substrate was cleaned with acetone, methanol, hydrochloric acid and purified water. After the substrate was inserted into the MBE growth chamber, (1) the substrate was thermally cleaned at 750 °C for 10 min. (2) Then, a thin GaN layer was deposited at 650 °C for 3 min. (3) Next, InN layer was grown at 435 °C for 30 min. (4) The InN layer was then irradiated with N radical beam at 435 °C with a N₂ gas supply of 2.0 sccm and plasma power of 200 W for 60 min. Steps (3) to (4) were then repeated. Finally, InN film was regrown on the irradiated template at 435 °C for 60 min. The growth rate was about 450 nm/h and the resultant total thickness of the sample was ~900 nm as shown in Fig. 2.

![Fig. 1 Growth time chart of the InN sample with two repetitions of N radical irradiation on the InN template.](image)

After the growth, TEM has been used to study dislocation behavior in the irradiated InN layers and the regrown InN film. Cross-sectional view specimens with electron beam incidence parallel to [0002] of InN have been prepared by focused ion beam etching (HITACHI FB-2100). A TEM (JEOL JEM2010) with an accelerating voltage of 200 kV was used in these studies. The samples were characterized using bright field observation with diffraction vectors, \( g = [1100] \) and \( g = [0002] \) to determine the dislocations with edge-type and screw-type components, respectively. The average dislocation density was measured from TEM micrographs with taking ~100 nm thickness for electron beam transparent region into account.

**3 Results and Discussions**

Figure 3 shows representative cross-sectional TEM images of the 900 nm thick InN grown on the MOCVD-grown (0001) GaN/sapphire substrate. The thickness of the first irradiated InN layer was ~225 nm, the second irradiated InN layer was ~225 nm, and the top regrown InN layer was 450 nm, as estimated from the TEM images. Note that threading dislocations are the only extended defect that grow through to the free surface of the InN layer. The threading dislocation density is in the order of \( 10^{10}/\text{cm}^2 \) although that of MOCVD-grown (0001) GaN template is in the order of \( 10^{9}/\text{cm}^2 \). It is generally known that c-plane InN and c-plane GaN have large difference in lattice constant of about 11%. This is the main reason of the high threading dislocation density in the grown InN layer. Generally, the Burgers vectors of edge dislocation, screw dislocation, and mixed dislocation in InN are defined as \( b = \frac{1}{3} < 11\bar{2}0 > \), \( b = \frac{1}{3} < 0001 > \), and \( b = \frac{1}{3} < 11\bar{2}3 > \),
respectively. In the c-plane InN growth, most threading dislocations in the InN film are found to be edge dislocation. At present, the common density of screw dislocation and edge dislocation in InN are around $2 \times 10^{9}$ cm$^{-2}$ and $2 \times 10^{10}$ cm$^{-2}$, respectively. Figure 3(a) shows the TEM image with a diffraction vector, $g = [\bar{1}1\bar{0}]$. Threading dislocations observed in this figure should have the edge component. As can be seen, a high density of threading dislocations was generated at the interface of GaN and InN, and these dislocations propagated into the InN layers. It is important to realize that these dislocations were clearly bent and some of them merged at the regrowth interfaces. The density of the edge dislocations in the first irradiated InN layer was estimated to be about $2.8 \times 10^{10}$ cm$^{-2}$. The dislocation density reduced to about $2.0 \times 10^{10}$ cm$^{-2}$ in the second InN irradiated layer, and then to about $1.3 \times 10^{10}$ cm$^{-2}$ in the top regrown InN layer. This means that the two stages of threading dislocation density reduction can be clearly observed. It should be noticed that, however, effect of repeating in situ surface modification by N radical irradiation on the threading dislocation density reduction was less than the results in ref. 19. The difference might be caused by different film thickness of InN template before in situ surface modification. They are 225 nm and 450 nm for this study and ref. 19, respectively. Threading dislocation density and/or surface morphology of InN can be changed as increasing the film thickness. Therefore, as we discuss later, optimization of the film thickness of InN template for in situ surface modification should be considered.

Figure 3(b) shows the TEM image with a diffraction vector, $g = [00\bar{2}]$. Threading dislocations observed in this image should have the screw component. As can be seen, a very few screw dislocations generated at the interface of GaN and InN as indicated by white arrows in Fig. 3(b). As a result, we found that both irradiated InN layers and regrown InN layer have low screw dislocation density, around $3 \times 10^{9}$ cm$^{-2}$. However, compare to our previous result, the screw dislocation was slightly bent at the regrowth interfaces and a few additional screw dislocations was generated at the interface of irradiated InN layers and regrown InN layer. From the TEM observation results, clearly, repeating the N radical irradiation process on the InN template properly could further reduce threading dislocation density in the top regrown InN layer.

In addition, another experiment with five repetitions of N radical irradiation process have also been carried out. In this experiment, in order to keep the total thickness of the sample constant at $-900$ nm, the thickness of each irradiated InN layer was reduced to $-90$ nm and the top regrown InN layer was kept constant at $-450$ nm as shown in Fig. 4. Figure 5 shows a cross-sectional TEM image of the sample. As can be seen, the interfaces between each irradiated layers and the regrown layer could not be identified. Because of large lattice mismatch between InN and GaN, three-dimensional island growth of InN started on GaN template, and flat surface was formed by coalescence of the island as film thickness increased. In this experiment, a streak pattern in RHEED monitoring was not observed after the first InN template growth with 90 nm thickness, indicating that the flat surface was not obtained starting from the second layer of the InN template. Due to the repetition of N radical irradiation against the rough InN surface and thin InN growth on top of that, as a consequence, the flat surface could not be obtained at the top regrown InN layer. From this result, it is suggested that in order to make use of the N radical irradiation process effectively, it is necessary to optimize the thickness of each InN layer during the N radical irradiation process.

Overall, the presented results show that the repeatability of this method to reduce threading dislocation density in InN with the optimum thickness of the template was confirmed as shown in Fig. 5. The mechanism of the threading dislocation density by this method are discussed from the following points. Generally, dislocation lines may end only at free surfaces, high-angle grain boundaries, or other dislocations, and threading dislocation densities are reduced by either annihilation of threading segments with antiparallel Burgers
vectors or by reactions in which two threading dislocations combine to form one threading dislocation[24]. This can be achieved through various paths such as bending at the surface, merging of dislocations, and interaction with other defects. When we focus on the in situ surface modification by radical beam irradiation growth method, firstly, dislocation bending can be included because of the presence of the interface between the irradiated InN template and the regrown InN region. To date, it is well known that faceted surfaces are effective for threading dislocation bending at the interfaces in III-nitrides semiconductors[25,28]. When compare with our proposed method, the faceted surface morphology was not obtained on the irradiated InN template. On the other hand, the surface changed from atomically flat surface to three-dimensional rough surface[9]. The surface morphological changes by N radical irradiation have also been reported by another group[27,30]. Cantu et al. reported that the surface roughness of the stressed layer during growth helped diminish the energy barrier during the initial stage of threading dislocation inclination and illustrated threading dislocation inclination due to the rough surfaces[90]. It is considered that changes in surface roughness introduced by in situ N radical irradiation can be effective for the inclination of threading dislocations in regrown InN.

Secondly, the inclination of the threading dislocations may lead to merging of dislocations. These merging of dislocations will be enhanced when a film contains a high density of dislocations. Since threading dislocation density in InN template is high which is around \(10^{19}\ \text{cm}^{-2}\) [15,16,30,31], merging of dislocations at the interface of the irradiated InN template and the regrown InN film could play the dominant role in the reduction of the dislocation density. We have confirmed by TEM that some dislocation merged at the regrowth interface and resulted in one single dislocation [39].

When we regrew InN on InN template which was irradiated with a higher plasma power (600 W), the threading dislocation reduction was more obvious compared to that of 200 W, as shown in Fig. 6 indicated by white arrows. We found that after the N radical irradiation with a higher plasma power, a higher carrier density in the InN template was confirmed by Hall effect measurement. This might be the result of increased point defects due to the higher plasma power during the N radical irradiation[25,33]. Since the dislocation motion is strongly affected by the interaction with point defects[34], we also consider the interaction between threading dislocations with point defects might be one of the threading dislocation reduction mechanisms in this method.

### 4 Conclusion

In this study, the repeatability of in situ surface modification by radical beam irradiation to reduce threading dislocation density in InN film was investigated. TEM observation showed evidence that the threading dislocation density was reduced from \(2.8\times10^{10} \text{cm}^{-2}\) in the first irradiated layer to \(2.0\times10^{10} \text{cm}^{-2}\) in the second irradiated layer, and to \(1.3\times10^{10} \text{cm}^{-2}\) in the top regrown InN film. This means that the two stages of threading reduction can be clearly observed.

Further repetitions of N radical irradiation process should be carried out with the optimum thickness of the template in order to study the maximum potential of threading dislocation density reduction by this method. It is assumed that the reduction of threading dislocation obtained in this study was resulted from dislocations bending at the regrowth interface, merging of dislocations and probably interaction with other defects.

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