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Characterization of pit formation in III-nitrides grown by metalorganic chemical vapor deposition

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Pit formation in III-nitride heterostructures such as InGaN/GaN and AlGaN/GaN grown by metalorganic chemical vapor deposition was characterized by transmission electron microscopy. The pit formation related with V defects has been reported in the InGaN/GaN multiple quantum well with high In composition [Appl. Phys. Lett. 79, 215 (2001)]. In this letter, we found that the mechanism of pit formation strongly depends on the indium and aluminum compositions in In\(_x\)Ga\(_{1-x}\)N and Al\(_x\)Ga\(_{1-x}\)N layers, respectively. By increasing the indium composition, the origin of pits is changed from the vertex of threading dislocations to the stacking mismatch boundaries induced by stacking faults and the three-dimensional island growth at the initial stage due to the large lattice mismatch. By increasing the aluminum composition, the origin of the pits also varied from the surface undulation due to the elastic misfit strain to the vertex of threading dislocations. In addition, several inversion domains observed in III nitrides result in pit formation in the surface of films. © 2002 American Institute of Physics.

Nitride alloys (GaN, AlGaN, and InGaN) are promising semiconductors which have applications in both electronic devices operating at high temperature, high frequency, and high power and optical devices including light-emitting diodes in the blue-green and ultraviolet wavelength region.\textsuperscript{1,2} For In\(_x\)Ga\(_{1-x}\)N and Al\(_x\)Ga\(_{1-x}\)N layers grown on GaN, an increase in the indium (In) and aluminum (Al) compositions may inevitably cause misfit strains in films. Beyond a critical thickness, several structural defects such as misfit dislocations, stacking faults, V-shaped pits, etc., are generated by plastic relaxation.\textsuperscript{3} However, also at smaller thicknesses, elastic relaxation may be induced by the formation of islands, surface undulation, etc., as observed in InGaP/InAsP, In\(_{0.77}\)Ga\(_{0.23}\)N/InGaAs, SiGe/Si, and so on.\textsuperscript{3,4,5,6}

Recently, we reported two models of the formation of V-shaped pits in InGaN/GaN multiple quantum wells (MQWs) with high In composition.\textsuperscript{7} Also, the undesirability and the effect on the emission properties of these pits were previously demonstrated by transmission electron microscopy (TEM), cathodoluminescence, and photoluminescence.\textsuperscript{3,5,8,9} For the suppression of pit formation, therefore, a clear understanding of the mechanisms of pit formation is needed and the microstructure of the pits observed in various III-nitride films grown on sapphire substrate must be evaluated in detail. In this work, we investigated the pit formation originating from different sources in InGaN/GaN and AlGaN/GaN heterostructures with various In and Al compositions using TEM.

All samples were grown on c-plane sapphire substrates with a nominal 25 nm thick GaN nucleation layer by a horizontal metalorganic chemical vapor deposition reactor operating at low pressure. Trimethylgallium, trimethylindium, trimethylaluminum, and ammonia were used as the source precursors for Ga, In, Al, and N, respectively. In order to analyze the effect of the strain on the mechanism of pit formation in III-nitrides, thick In\(_x\)Ga\(_{1-x}\)N layers (~800 Å) were grown at temperatures ranging from 830°C (x\(_{\text{In}}\) = 35%) to 880°C (x\(_{\text{In}}\) = 10%) and thick Al\(_x\)Ga\(_{1-x}\)N layers (~1000 Å) were grown at 1040°C after growing the Si-doped underlying GaN layer of ~2 μm thickness at 1080°C. The In and Al compositions in the In\(_x\)Ga\(_{1-x}\)N and Al\(_x\)Ga\(_{1-x}\)N layers were 10%, 13%, 19%, and 35% and 16%, 25%, 44%, and 54%, respectively.

In order to characterize the pit formation of III-nitrides, TEM specimens were prepared in cross section along [1120] zone axis using Tripod mechanical polishing followed by low temperature Ar ion milling at 4.5 kV in a Gatan Duomill 660 DIF with sector speed control. The ion energy was gradually reduced during the final stages of thinning to minimize the surface damage of samples. Bright-field (BF) images and high-resolution TEM (HRTEM) images were recorded on a JEOL JEM-2000EX TEM at 200 kV and JEOL JEM-3010EX at 300 kV.

Figure 1 shows the cross sectional BF TEM micrographs from the In\(_x\)Ga\(_{1-x}\)N layers grown with various In compositions on the GaN buffer layer. The theoretical critical In composition on the formation of the misfit dislocation of the ~800 Å In\(_x\)Ga\(_{1-x}\)N grown on GaN layers is less than 5%.

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below which the InGaN layers show coherent growth.\textsuperscript{10} Therefore, we expect the In composition of all In\textsubscript{\textit{x}},Ga\textsubscript{1\textit{x}}N layers studied here to have a value more than the critical In composition on the formation of the misfit dislocation. For In\textsubscript{0.1},Ga\textsubscript{0.9}N/GaN, no structural defects such as misfit dislocations, stacking faults, pits, etc., were observed within the In\textsubscript{0.1},Ga\textsubscript{0.9}N layer. With a further increase in the In composition of the In\textsubscript{\textit{x}},Ga\textsubscript{1\textit{x}}N layer, the V-shaped pits are generated to relax the stored misfit strain, as reported previously.\textsuperscript{8,11} It has been reported that a V-shaped pit is always connected with a threading dislocation from the GaN buffer layer at the bottom and only a small fraction of the threading dislocations causes the formation of the V-shaped pits in the InGaN layers and the InGaN/GaN MQWs.\textsuperscript{8,11} However, the density of V-shaped pits in the In\textsubscript{0.13},Ga\textsubscript{0.87}N layer in Fig. 1(c) has a larger value than the threading dislocation density in the GaN buffer layer, which indicates that the formation of V-shaped pits in the InGaN with high In composition has a different origin, as reported previously for the InGaN/GaN MQW systems.\textsuperscript{7} We reported that in the InGaN/GaN MQWs with high In composition (\textsim 30\% In composition), most of the V-shaped pits are generated from stacking mismatch boundaries induced by stacking faults.\textsuperscript{7} Although the In \textsubscript{0.19},Ga\textsubscript{0.81}N/GaN used here has an In composition of less than 20\%, the part of V-shaped pits originate from stacking mismatch boundaries induced by stacking faults due to the increased InGaN layer thickness [Fig. 2(a)] compared to the InGaN/GaN MQW. For In\textsubscript{0.35},Ga\textsubscript{0.65}N/GaN, V-shaped pits with a density of more than $10^{10}$ cm\textsuperscript{-2} are observed on the surface. Unlike the pits in the In\textsubscript{0.19},Ga\textsubscript{0.81}N/GaN sample, the In\textsubscript{0.35},Ga\textsubscript{0.65}N regions with flat (0001) top surfaces between pits have a large number of stacking faults and stacking mismatch boundaries formed at the initial growth stage of InGaN [Fig. 2(b)], that is, columnar structures including stacking faults. To relax the large lattice mismatch between In\textsubscript{0.35},Ga\textsubscript{0.65}N and GaN, the initial In\textsubscript{0.35},Ga\textsubscript{0.65}N layer was grown with the morphology of three-dimensional (3D) faceted islands.\textsuperscript{12} A high density of stacking faults is clearly seen in the islands and the boundaries between subgrains by islands are visible like the growth of a GaN nucleation layer grown on sapphire substrate.\textsuperscript{13} Therefore, we think the pits in the In\textsubscript{0.35},Ga\textsubscript{0.65}N/GaN sample are caused by such a 3D growth due to the lattice misfit at the initial growth of the InGaN layer.

As a result, we classified the origin of pit formation in the In\textsubscript{\textit{x}},Ga\textsubscript{1\textit{x}}N grown on the GaN buffer layer into three models as shown in Figs. 4(a), 4(b), and 4(c). For the In\textsubscript{\textit{x}},Ga\textsubscript{1\textit{x}}N layer with low In composition, pits are generated at the vertex of threading dislocations [Fig. 4(a)]. For the In\textsubscript{\textit{x}},Ga\textsubscript{1\textit{x}}N layer with medium In composition, pits are mainly generated from the stacking mismatch boundaries induced by stacking faults [Fig. 4(b)]. A further increase in In composition of the In\textsubscript{\textit{x}},Ga\textsubscript{1\textit{x}}N layer results in the 3D island growth at the initial stage and pit formation at the continuous growth [Fig. 4(c)].

To investigate the effect of Al composition on pit formation, cross sectional BF TEM micrographs from the Al\textsubscript{\textit{x}},Ga\textsubscript{1\textit{x}}N layers grown with various Al compositions on the GaN buffer layer were obtained as shown in Fig. 3. The theoretical critical Al composition on the formation of the misfit dislocation in the \textsim 1000 Å Al\textsubscript{\textit{x}},Ga\textsubscript{1\textit{x}}N layer grown on GaN layers is less than 15\%.\textsuperscript{10} No pits are detected in the Al\textsubscript{\textit{x}},Ga\textsubscript{1\textit{x}}N layers with up to 25\% Al composition. Only stacking faults and cracks in the AlGaN layers are observed due to the misfit strain and the thermal coefficient difference, respectively (not shown). For the Al\textsubscript{0.44},Ga\textsubscript{0.56}N/GaN sample, however, the surface of the Al\textsubscript{0.44},Ga\textsubscript{0.56}N layer shows many pits that are not associated with structural defects such as threading dislocations, stacking faults, and so on [Figs. 3(b) and 3(d)]. The size of these pits is very small, an average diameter of 7 nm and an average height of \textsim 3 nm, compared to that of pits in InGaN layers grown on GaN.\textsuperscript{7,8,10} It is believed that the formation of pits in Al\textsubscript{\textit{x}},Ga\textsubscript{1\textit{x}}N/GaN can be ascribed to the surface undulation by the increased elastic misfit strain as observed in III–V and II–VI semiconductor

**FIG. 1.** Cross sectional BF TEM images using \textbf{g} = 0002 from the In\textsubscript{\textit{x}},Ga\textsubscript{1\textit{x}}N layers grown with the In composition of (a) 10\%, (b) 13\%, (c) 19\%, and (d) 33\% on the GaN buffer layer. The dashed lines indicate the In\textsubscript{\textit{x}},Ga\textsubscript{1\textit{x}}N/GaN interface.

**FIG. 2.** (a) HRTEM image obtained from the dashed rectangle in Fig. 1(c). The observed V-shaped pit has a stacking fault on (0001) planes in its lower position. (b) HRTEM image obtained from the In\textsubscript{0.35},Ga\textsubscript{0.65}N on the GaN buffer layer. The observed V-shaped pit has a stacking fault on its lower plane. (c) HRTEM image obtained from the In\textsubscript{0.35},Ga\textsubscript{0.65}N/GaN interface.

**FIG. 3.** Cross sectional BF TEM images using \textbf{g} = 0002 from the Al\textsubscript{\textit{x}},Ga\textsubscript{1\textit{x}}N layers grown with the Al composition of (a) 16\%, (b) 44\%, and (c) 54\%. (d) HRTEM image obtained from the dashed rectangle in (b). The dashed lines indicate the Al\textsubscript{\textit{x}},Ga\textsubscript{1\textit{x}}N/GaN interface.
The density of pitting is more than \(10^{10} \, \text{cm}^{-2}\) in the surface. On the other hand, misfit dislocations are generated at the interface of the Al\(_{0.54}\)Ga\(_{0.46}\)N/GaN sample in order to plastically relax the stored misfit strain as shown in Fig. 3. Also, V-shaped pits associated with threading dislocations at the their bottoms are observed like InGaN/GaN heterostructures. These V-shaped pits have an open hexagonal, inverted pyramid with \(\{10\overline{1}1\}\) sidewalls. 8,10

In consequence, we classified the origin of the pit formation in the Al\(_{x}\)Ga\(_{1-x}\)N layer on the GaN buffer layer into two models as shown in Figs. 4(a) and 4(d). For the Al\(_{x}\)Ga\(_{1-x}\)N layer with \(\sim 40\%\) Al composition, pits are generated by the surface undulation due to the stored elastic misfit strain [Fig. 4(d)]. For the Al\(_{x}\)Ga\(_{1-x}\)N layer with more than \(\sim 50\%\) Al composition, however, pits are mainly generated at the vertex of threading dislocations [Fig. 4(a)] like InGaN/GaN.

Figure 5 is the cross-sectional BF TEM micrographs showing the origins of pitting from inversion domains observed in various III-nitride heterostructures. Figures 5(a) and 5(b) demonstrate that IDBs generated from the GaN/substrate interface lead to formation at the surface. Unlike the pit formation by threading dislocations, 8,10 however, the pit formation by IDBs does not disrupt the InGaN/GaN MQWs as shown in Fig. 5(a). The IDs observed near the surface area of nitride films with the AlGaN (Mg-doped)/GaN superlattice also result in the pit formation due to the different growth rates within IDs compared to matrix around them as reported previously [Fig. 5(c)]. 14,15

In summary, the pit formation in In\(_{x}\)Ga\(_{1-x}\)N and Al\(_{x}\)Ga\(_{1-x}\)N layers with various In and Al compositions grown on the GaN buffer layers was studied using TEM. We found that pits were formed from various origins such as threading dislocations, stacking mismatch boundaries, 3D island growth, surface undulation, and IDs depending on In and Al compositions.

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