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Observations of Segregation of Al in AlGaN Alloys

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Transmission electron microscopy has been used to characterize Al segregation in $Al_{0.1}Ga_{0.9}N$ and $Al_{0.3}Ga_{0.7}N$ alloys grown by metal organic chemical vapor deposition on 6H-SiC. It has been found that an interlayer of AlGaN alloy with much higher Al content was formed at first, followed by normal growth of nominal composition of AlGaN alloy. In $Al_{0.1}Ga_{0.9}N$ and $Al_{0.3}Ga_{0.7}N$ films, dislocation lines were also found to have more Al segregated than those regions free of dislocations in the matrix. Furthermore, it shows that more Al atoms segregate to an edge dislocation than to a screw one.

Introduction AlGaN is a key material for optoelectronic and microwave device application [1, 2]. The microstructures of AlGaN layers have been a subject of intensive research for the past few years [2–4]. AlGaN grown on 6H-SiC substrate often contains a large number of dislocations (dislocation density $\sim 10^9$ cm^{-2}) due to the lattice mismatch which depends on the Al composition in the alloys. A lower density of dislocations is necessary for high quality GaN films as the dislocations degrade the optoelectronic properties. Structural characteristics of dislocations in GaN related alloys with their effects on the optoelectronic properties have been characterized in the past [5–9]. However, composition variation within AlGaN ternary alloys has not been noticed. In this paper, we present evidence of Al segregation in the initial stage of deposition and around the dislocations in the grown films of AlGaN alloys based on the observations of transmission electron microscopy (TEM) with X-ray energy dispersive spectroscopy (EDX) in a nanometer spatial resolution [10].

Experimental Conditions 400 nm thick AlGaN films in two different compositions were grown on 2 inch n-type 6H-SiC (0001) substrates by metal organic chemical vapor deposition (MOCVD). The nominal concentrations of Al and Ga were deduced from double-crystal X-ray diffraction to be $Al_{0.1}Ga_{0.9}N$ and $Al_{0.3}Ga_{0.7}N$. Both the AlGaN films were doped with Si in the order of 10^{18} cm^{-3}. The deposition temperatures were in the range of 1050–1250 °C. The source materials used were triethylaluminium, triethylgallium, and NH_3. The cross-sectional TEM specimens were prepared by a conventional sandwiched method consisting of gluing, mechanical thinning, and ion milling by Ar ion beam to perforation. TEM was carried out in a JEOL JEM 2010F microscope with a field-emission gun which can form an electron probe in a 0.5 nm diameter size. The operating voltage was set at 200 kV. The compositions were obtained by EDX from an Oxford Instrument EDX detector with an ultrathin window. All necessary pre-

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cautions for the quantitative analysis of compositions had been taken before the acquisition of the spectra [10, 11]. The beam broadening size is estimated to be less than 2 nm. The concentrations were determined by assuming that the nitrogen concentration is constant, and the sum of Ga and Al are 100% in total.

Results and Discussion Figure 1[2]) shows the TEM images of $Al_{0.1}Ga_{0.9}N$ and $Al_{0.3}Ga_{0.7}N$ samples. At the interface with SiC substrate, it can be seen that there is an interlayer in dark contrast (bright field images of Figs. 1a and c), which has a different structure from the above AlGaN film as shown in the high-resolution images (Figs. 1b and d). EDX results (Table 1) reveal that the interlayer has a composition of $Al_{0.4}Ga_{0.6}N$ for deposition of the $Al_{0.1}Ga_{0.9}N$ alloy and $Al_{0.8}Ga_{0.2}N$ for that of the $Al_{0.3}Ga_{0.7}N$ alloy, respectively. Similar phenomena with Al-rich phases have been reported as a result of phase separation [12–14]. The formation of the interlayer suggests that nucleation on the substrates with higher concentration of Al may reduce lattice mismatch with SiC. After the nucleation stage, the alloy growth proceeds with nominal composition. The formation of Al-rich interlayer may affect the dislocation density in the film grown because of their lattice mismatch between them.

The dislocations grown from the interface between AlGaN and SiC are clearly seen in Fig. 1. The mean dislocation spacing is about 100 nm (the density $\sim 9 \times 10^9$ cm^{-2}) in

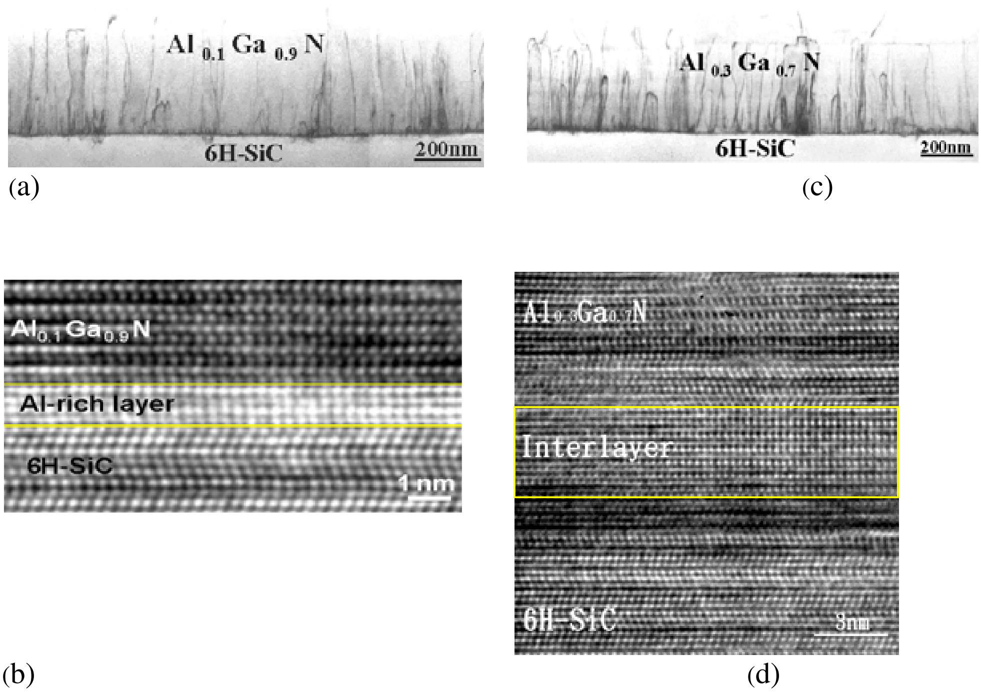

Fig. 1 (colour). Bright field and high-resolution TEM images of a), b) $Al_{0.1}Ga_{0.9}N$ and c), d) $Al_{0.3}Ga_{0.7}N$

[2]) Colour figure is published online (www.physica-status-solidi.com).

Table 1

Al compositions (in at%) measured from EDX. The bulk values were obtained from an electron beam in micron size. The rest values were obtained from nano beam measurements

buffer layer	$Al_{0.1}Ga_{0.9}N$	$Al_{0.3}Ga_{0.7}N$
bulk	10.1	29.0
interlayer	46.9	79.3
matrix	9.6	21.0
edge	19.1	36.0
screw	12.6	30.9
mixed	14.2	33.8

the $Al_{0.1}Ga_{0.9}N$ film and about 40 nm (the density $\sim 5 \times 10^{10}$ cm^{-2}) in the $Al_{0.3}Ga_{0.7}N$ film. The edge type dislocations have the Burgers vector 1/3 $[11\bar{2}0]$, the screw [0001], and the mixed 1/3 $[11\bar{2}3]$. Detailed examination reveals that the dark line of contrast in the TEM images is mainly due to the strain field of a dislocation rather than a precipitate. EDX results from various areas probed by micron-size and nanometer-size electron beams are given in Table 1. The regions in the films probed by a large beam have a bulk composition close to the nominal value. However, the results from nanobeam probe show that the matrix free of dislocations has lower concentration of Al. In contrast, dislocations in both alloys have more Al segregated with. The segregation level of Al is highest at edge type dislocations, followed by the mixed type and the screw type ones. For the $Al_{0.1}Ga_{0.9}N$ case, the Al concentration at the edge dislocations is enriched about two times as high as that in the matrix. In $Al_{0.3}Ga_{0.7}N$, the Al enriched factor at the edge dislocations is about 1.5 times compared with the value in the matrix free of dislocations. The Al composition profiles across the different dislocations in the $Al_{0.1}Ga_{0.9}N$ alloy are demonstrated in Fig. 2. It is apparent that Al is enriched around the dislocation cores, but depleted at both sides in 2–3 nm regions. The depletion of Al to 4–8 at% is very significant in $Al_{0.1}Ga_{0.9}N$. Similar behaviors of Al segregation around dislocations are observed in the $Al_{0.3}Ga_{0.7}N$ alloy as well.

It is known that the Al atom has a smaller size than the Ga one. Therefore, Al segregation to the dislocations in AlGaN alloys is reasonable because the strain energy

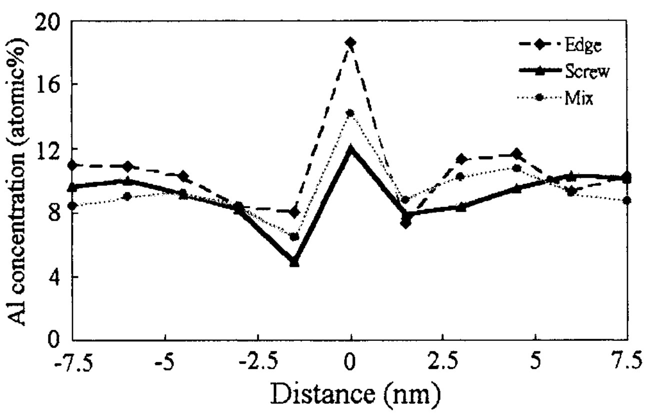

Fig. 2. Al concentration profiles across edge, screw, and mixed dislocations in $Al_{0.1}Ga_{0.9}N$

of dislocation can be reduced. It has been shown that the line energy of edge dislocation is higher than that of screw in GaN [15]. However, the enrichment of Al at an edge dislocation appears much stronger than at a screw dislocation from our EDX results. This may imply that the interaction energy of Al atoms with an edge dislocation is larger than with a screw one, probably due to the absence of elastic displacements parallel to the basal plane for the screw [16]. The reason about the Al depletion surrounding the dislocation cores is not clearly known. The presence of Al segregation around dislocations may pin the dislocation lines, which makes them difficult to move. As a result, the dislocation densities in AlGaN alloys may not be easily reduced. Thus, it is necessary to change the deposition conditions if a low dislocation density is desired for the AlGaN layers.

Conclusions Al segregation in $Al_{0.1}Ga_{0.9}N$ and $Al_{0.3}Ga_{0.7}N$ alloys grown on 6H-SiC has been observed by TEM. An Al-rich interlayer with 47 and 80 at% was formed before normal growth of the film during deposition of $Al_{0.1}Ga_{0.9}N$ and $Al_{0.3}Ga_{0.7}N$, respectively. The Al concentration enriched at the dislocation core can be about two times as high as that in the matrix. The increase of Al concentration at the dislocations strongly depends on the dislocation type, which is the highest for the edge type and the next for the mixed type, followed by the screw type.

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