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Influence of Mg doping on structural defects in AlGaN layers grown by metalorganic chemical vapor deposition

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Influence of Mg doping on structural defects in $Al_{0.13}Ga_{0.87}N$ layers grown on sapphire substrates by metalorganic chemical vapor deposition were studied using transmission electron microscopy. By increasing the Mg source flow rate, the reduction of dislocation density occurred up to the Mg source flow rate of 0.103 μmol/min. While the vertical type inversion domain boundaries (IDBs) were observed in the $Al_{0.13}Ga_{0.87}N$ layers grown with the low Mg source flow rate, the IDBs in the $Al_{0.13}Ga_{0.87}N$ layers grown with the high Mg source flow rate have horizontally multifaceted shapes. The change of polarity by the IDBs of horizontal type also resulted in the 180° rotation of pyramidal defects within the same AlGaN layer.
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Wurtzite GaN-based (GaN, InGaN, and AlGaN) semiconductors are currently under enormous investigation as promising materials for optoelectronic, high-temperature, and high-power devices due to some properties such as large direct band gap, high thermal stability, and strong interatomic bonds.[1] For good photon and current confinement, a low aluminum (Al) composition AlGaN cladding layer is used in the actual InGaN/GaN multiple quantum well (MQW) light emitting diode (LED) and laser diode (LD) structures.[2] Also, AlGaN layers as the first layer grown on sapphire substrates without underlying GaN layers, which can avoid the generation of cracks, can be applied as wide-band-gap windows and buffer layers in AlGaN/GaN optoelectronic devices.[3] However, the crystalline quality of AlGaN is degraded due to the small diffusion length of Al atoms on the surface of the low temperature nucleation layers.[4] That is, the smaller island size by low diffusion length results in the increase of a threading dislocation density.

Planar defects such as stacking fault, inversion domain boundary (IDB), and stacking mismatch boundary are observed in GaN layers grown on sapphires, in addition to threading dislocations.[5,6] Two typical types of IDBs have been observed in GaN layers and AlGaN/GaN superlattice structures grown on sapphire substrates.[7–9] First, the vertical type IDBs lying in the $\{10\bar{1}0\}$ planes have a translation through a distance of $c/2$ along the [0001] direction to avoid wrong bonds. The $\{10\bar{1}0\}$ IDBs start at the interface with the sapphire substrate, propagate from the GaN/sapphire interface to the top surface of the sample, and induce the different growth rate around IDBs. Second, the horizontal type IDBs lying in the {0001} and $\{11\bar{2}3\}$ planes are observed in the Mg-doped GaN layer grown on molecular-beam epitaxy (MBE).[8,9] The polarity of films is inverted at these domains. It has been reported that the control of the polarity greatly affects the optical and structural properties of LED and LD devices.[10,11] Therefore, the study of the IDBs in the Mg doped AlGaN layers must be scrutinized in detail. In this work, we investigated the effect of Mg doping in $Al_{0.13}Ga_{0.87}N$ layers on structural defects such as dislocation density, IDB, and pyramidal defects using transmission electron microscopy (TEM).

Mg-doped AlGaN layers were grown on *c*-plane sapphire substrates in a horizontal metalorganic chemical vapor deposition (MOCVD) reactor operating at low pressure. Trimethylgallium, trimethylaluminum, ammonia, and biscyclopentadienyl-magnesium (Cp_2Mg) were used as the source materials for Ga, Al, N, and Mg, respectively. The AlGaN epilayers were grown at 1100 °C after growing the GaN nucleation layers of about 25 nm thickness at 560 °C under the same growth conditions except the Cp_2Mg flow rate in $Al_{0.13}Ga_{0.87}N$ layers. The Cp_2Mg flow rate in $Al_{0.13}Ga_{0.87}N$ was changed from 0 μmol/min (undoped) to 3.172 μmol/min. The Al composition of AlGaN layers was obtained by high-resolution x-ray diffraction (HRXRD) measurement using Vegard's law.

For the cross-sectional TEM study, samples were mechanically polished into a wedge-shape using a Tripod polisher. The mechanically polished samples were finally ion milled. The prepared TEM specimens with the thickness of ~300 nm were examined in a JEOL JEM 2000EX microscope operating at 200 kV with a point resolution of 0.21 nm.

Figure 1 shows the HRXRD results of samples with various Cp_2Mg flow rates using the ω-mode scan of (0002) and $(1\bar{1}02)$ reflecting planes. It has been reported that Mg concentration shows a linear dependence on the Cp_2Mg flow rate.[12,13] As shown in Fig. 1, the full width at half maximum (FWHM) decreases upon increasing the Cp_2Mg flow rate,

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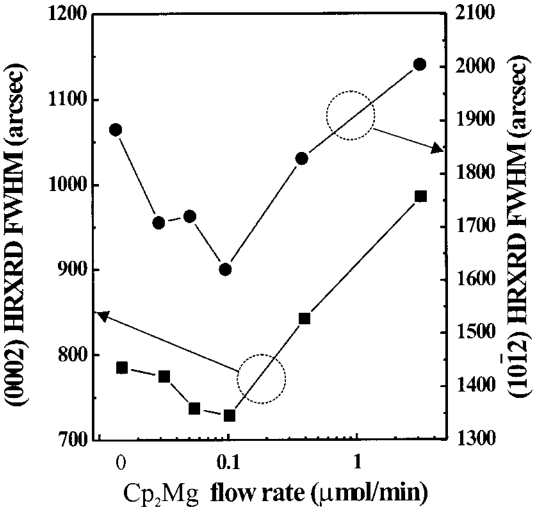

FIG. 1. Variation of the FWHM values of the HRXRD rocking curves for the (0002) and $(1\bar{1}02)$ reflections as a function of the Cp$_2$Mg flow rate of AlGaN layers.

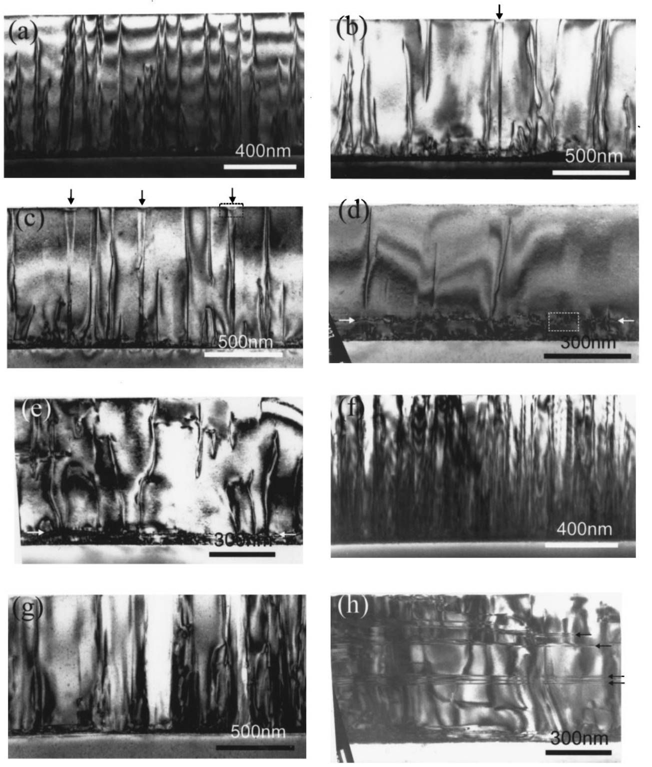

FIG. 2. Cross-sectional bright-field TEM images using $g=0002$ two beam of samples grown with the Cp$_2$Mg flow rate of (a) 0 μmol/min, (b) 0.055 μmol/min, (c) 0.103 μmol/min, (d) 1.031 μmol/min, and (e) 3.172 μmol/min, and using $g=1\bar{1}00$ two beam of samples grown with the Cp$_2$Mg flow rate of (f) 0 μmol/min, (g) 0.103 μmol/min, and (h) 3.172 μmol/min.

reaches the minimum at the Cp$_2$Mg flow rate of 0.103 μmol/min, then increases again. The minimum FWHMs obtained for the (0002) and $(1\bar{1}02)$ diffraction planes were 728 and 1629 arcsec, respectively. Such an evident broadening of the FWHM of HRXRD measurement of the Al$_{0.13}$Ga$_{0.87}$N layers compared to GaN layers grown on low temperature (LT) GaN nucleation layers has already been observed.[14] AlGaN layers grown on the LT GaN nucleation layer have a lattice mismatch between AlGaN and GaN. Also, because Al atoms or Al-containing molecules may have a small diffusion length on the surface of the LT nucleation layer, the size of the AlGaN islands that appeared in the initial stage of the growth is small.[4] Therefore, it causes the AlGaN layer with high-density dislocations and rough surface. The symmetric (0002) HRXRD rocking curve is only broadened by screw and mixed dislocations[15] and TEM images [Figs. 2(a)–2(e)] using $g=0002$ two-beam conditions also show screw and mixed dislocations. The results of HRXRD and TEM are in good agreement, which means the reduction of dislocation density up to the Cp$_2$Mg flow rate of 0.103 μmol/min. The density of total threading dislocation decreases from $4\times10^9\ \mathrm{cm}^{-2}$ (undoped) to $1.3\times10^9\ \mathrm{cm}^{-2}$ (0.103 μmol/min). For the asymmetric $(1\bar{1}02)$ rocking curve, the value of FWHM decreased rapidly in the low Mg doping condition, which indicated the reduction of an edge dislocation density[15] as shown in Figs. 2(f) and 2(g). It has been reported that Mg acts as a surfactant,[16] which modifies the surface mobility of the chemical species due to the reduction of surface energy in films on mismatched underlying layers. Therefore, we expect that the low concentration Mg leads to the change of Al surface mobility, increases the size of the initial AlGaN islands, and finally decreases the dislocation density (increased grain size) in Al$_{0.13}$Ga$_{0.87}$N layers as shown in Fig. 2. However, an increase of the Cp$_2$Mg flow rate beyond 0.103 μmol/min induces the large broadening of the (0002) and $(1\bar{1}02)$ FWHM due to the increased strain by the incorporation of Mg atoms with a large covalent radius.[17] The investigation of the defect distribution of AlGaN layers shows no increase of dislocation density as shown in Figs. 2(d) and 2(e). However, the high density of stacking faults on (0001) planes was observed in samples with the Cp$_2$Mg flow rate of more than 0.103 μmol/min [Fig. 2(h)]. The stacking faults are indicated by black arrows in Fig. 2(h). These stacking faults result in the broadening of the FWHM of HRXRD measurement. In optical characterization of these samples, we also observed the intense intensity of the Mg related PL peak in samples with the Cp$_2$Mg flow rate of 0.055 and 0.103 μmol/min (not shown in this letter).

Romano *et al.*[8] and Ramachandra *et al.*[9] have shown that Mg doping changes the polarity of GaN layers grown by molecular beam epitaxy (MBE) from Ga to N-polarity, which means IDBs. We investigated the influence of the Cp$_2$Mg flow rate on the microstructure such as IDBs and pyramidal defects of Al$_{0.13}$Ga$_{0.87}$N layers in detail. IDBs are observed in samples with the Cp$_2$Mg flow rate of less than 0.103 μmol/min except the undoped Al$_{0.13}$Ga$_{0.87}$N. Black arrows [Figs. 2(b) and 2(c)] indicate vertical type IDBs which originate in the GaN nucleation layer and pass the whole Mg-doped AlGaN layer. However, no IDB of this type was observed in undoped, Si-doped (not shown), and high Cp$_2$Mg flow rate (beyond 0.397 μmol/min) AlGaN layers (Fig. 2). Compared to reported IDBs in GaN layers grown on sapphire substrates, the width of IDBs becomes broad as the thickness of AlGaN increases. As shown in Figs. 3(a) and 3(c), the faceted step on the surface is the top region of inversion domain in the AlGaN layers. In the $\langle 11\bar{2}0\rangle$ projection, the facet angle is nearly 52° with respect to the basal plane, which corresponds to the $\{20\bar{2}3\}$ planes. The faceted steps on the inversion domain resulted from the slow growth rate within inversion domains compared to matrix around

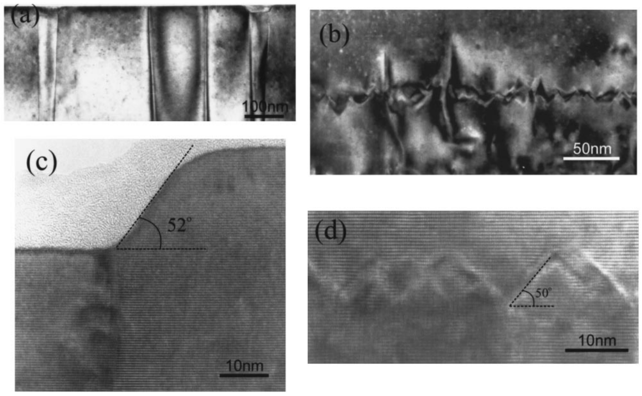

FIG. 3. High magnification images and high resolution TEM images of the IDBs of the samples grown with the Cp_2Mg flow rate of (a) and (c) 0.103 μmol/min and (b) and (d) 1.031 μmol/min, respectively. The magnified areas are indicated as dashed rectangles in Figs. 2(c) and 2(d).

them.[10] The exposure of Mg in the (0001) surface can cause an inversion of polarity from Ga to N polarity due to the formation of stable phases such as Mg_3N_2,[8,9] as reported by Ramachandra *et al.*[9] Since all samples were grown under the same growth conditions except the Cp_2Mg flow rate, therefore, the formation of vertical type IDBs in the low Mg source flow rate can probably be explained by the inversion of polarity in very small Mg-rich regions.

For the high Cp_2Mg flow rates (more than 0.397 μmol/min), in contrast, the different types of IDBs are observed in $Al_{0.13}Ga_{0.87}N$ layers. Bright field TEM images of AlGaN layers with high Cp_2Mg flow rates are shown in Figs. 2(d) and 2(e). The nearly horizontal IDBs are clearly visible, as indicated by the white arrows. High magnification TEM images [Figs. 3(b) and 3(d)] of the IDBs of horizontal type show multifaceted boundaries and no stacking faults around IDB. The facet angle ranges from 45° to 50° with respect to the basal plane, which is consistent with the result of Romano *et al.* for GaN layers grown by MBE on Ga-polarity (0001) templates.[8]

It has been observed that the shape of pyramidal defects having inclined $\{11\bar{2}3\}$ facets is very dependent on the polarity of film in Mg-doped GaN layers, that is, the tip of the pyramids always points toward the $[000\bar{1}]$ direction.[18] We investigated the shape of the pyramidal defect in $Al_{0.13}Ga_{0.87}N$ layers with high Cp_2Mg flow rates associated with multifaceted IDBs. It is commonly accepted that Ga-polarity GaN films are usually obtained by MOCVD growth,[19] which agrees well with our results. Figure 4 is magnified TEM images of a sample with the Cp_2Mg flow rate of 1.031 μmol/min. Pyramidal defects have an inverted shape in the HRTEM images shown above and below the multifaceted IDB, indicating that the change of polarity from Ga to N polarity in the high Mg-doped AlGaN layers induces the reverse shape of pyramidal defects within the same film. The growth of III-nitride films under Ga polarity has resulted in improved structural, electrical, and optical properties with smoother surface morphologies when compared to N-polarity growth.[10,11] Therefore, the control of polarity during the growth of Mg-doped AlGaN layers is very important in the case of high concentration Mg doping.

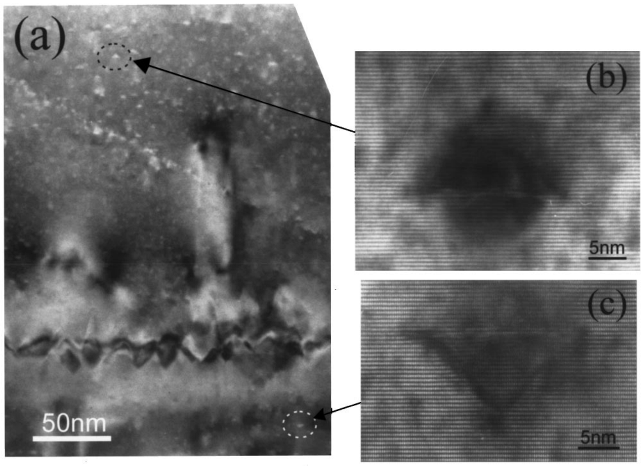

FIG. 4. (a) Cross-sectional bright-field TEM image of the multifaceted IDB of the sample grown with the Cp_2Mg flow rate of 1.031 μmol/min. (b) and (c) high resolution TEM images showing pyramidal defects with 180° rotation by the multifaceted IDB.

In summary, the structural defects such as dislocation, IDB, and pyramidal defects in Mg-doped AlGaN layers with various Mg source flow rates were studied using HRXRD and TEM. We found that the Mg source flow rate affects significantly the dislocation density, the type of IDBs, and the shape of pyramidal defects in AlGaN layers.

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