Structural characterization of $Al_{0.55}Ga_{0.45}N$ epitaxial layer determined by high resolution x-ray diffraction and transmission electron microscopy*

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Structural characteristics of $Al_{0.55}Ga_{0.45}N$ epilayer were investigated by high resolution x-ray diffraction (HRXRD) and transmission electron microscopy (TEM); the epilayer was grown on GaN/sapphire substrates using a high-temperature AlN interlayer by metal organic chemical vapor deposition technique. The mosaic characteristics including tilt, twist, heterogeneous strain, and correlation lengths were extracted by symmetric and asymmetric XRD rocking curves as well as reciprocal space map (RSM). According to Williamson–Hall plots, the vertical coherence length of AlGaN epilayer was calculated, which is consistent with the thickness of AlGaN layer measured by cross section TEM. Besides, the lateral coherence length was determined from RSM as well. Deducing from the tilt and twist results, the screw-type and edge-type dislocation densities are 1.0×10^{8} cm^{-2} and 1.8×10^{10} cm^{-2}, which agree with the results observed from TEM.

Keywords: $Al_xGa_{1-x}N$, high-temperature AlN interlayer, high resolution x-ay diffraction, transmission electron microscopy

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1. Introduction

The ternary $Al_xGa_{1-x}N$ alloys are of interest in the production of high-performance UV wavelength optoelectronic devices such as light-emitting diodes (LEDs) and photodetectors in the UV spectral region in the range of 200 nm–365 nm according to the band gap of AlGaN.[1,2] However, due to the large mismatch in lattice constant and thermal expansion coefficient between the epitaxial layer and substrates,[3–5] as well as the much lower mobility of Al versus Ga (Ga–N bond energy = 2.2 eV, Al–N bond energy = 2.88 eV),[6] AlGaN materials typically have high density defects, which act as non-radiative recombination centers. As a result, these high density defects are detrimental for device applications. Therefore, it is vital to conduct detailed structural analyses and improve crystal quality to fabricate efficient devices.

Heteroepitaxial thin films with large lattice mismatch to the substrate can be modeled by mosaic structure consisting of many small hexagonal grains,[7–10] as illustrated in Fig. 1.[11,12] Based on the mosaic model, the structural properties can be thoroughly analyzed by using Williamson–Hall plots calculated from XRD data, which provide information of the layers including tilt angle ($\alpha^{\rm tilt}$), heterogeneous strain ($\varepsilon_{\perp}$), vertical coherence length ($L_{\perp}$), and lateral coherence length ($L_{\parallel}$).[13] Meanwhile, a reciprocal space map (RSM) of asymmetric reflections was also performed. Both the mosaic tilt and lateral coherence length can also be determined from the inclination angle of asymmetric reciprocal lattice points (RLPs).[11] In addition, a mosaic twist can be extracted from rocking curves of asymmetric reflections in off-axis skew symmetric geometries with increasing lattice plane inclination.[14,15] Recently, some literature discussed the above mosaic model and estimated the structural quality of III-nitrides,[16–24] which proved the validity of this method.

In contrast, transmission electron microscopy (TEM) measurements are performed in order to obtain a direct observation of structural defects. Therefore, the aim of the present paper undertakes a detailed study on the structure characteristics of AlGaN layer, including tilt angle, twist angle, het-

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erogeneous strain, vertical coherence length, lateral coherence length, and dislocation (edge and screw dislocation densities). Furthermore, it demonstrates determining structural quality by different XRD measurement methods, which are verified by the results obtained by TEM.

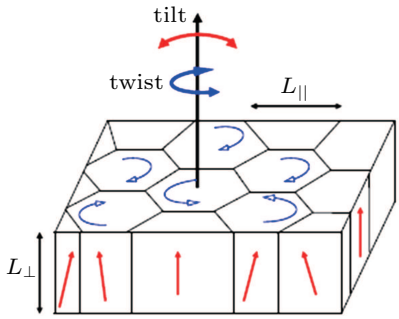


Fig. 1. (color online) Illustration of a mosaic structure with characteristic parameters.

2. Experimental procedures

$Al_{0.55}Ga_{0.45}N$ epilayer was grown at 1100 °C on GaN template with high temperature AlN layer (HT-AlN) as the interlayer at a reactor pressure of 76 Torr (1 Torr = 1.33322×10^2 Pa) by metal organic chemical vapor deposition (MOCVD). The molar flux of trimethylgallium (TMGa), trimethylaluminum (TMAl) and ammonia (NH_3) were 1.375×10^{-5} mol/min, 1.18×10^{-5} mol/min, and 1.62×10^{-1} mol/min, respectively. The detail of the growth procedure was described in Ref. [25]. The thickness of AlGaN layer was measured as about 0.502 μm by TEM.

The x-ray diffraction measurements were performed using high resolution x-ray diffractometer (PANalyticalX'Pert Pro MRD) equipped with a four-crystal Ge (220) mono chromator and a three reflection Ge (220) analyzer, delivering x-ray of Cu Ka line = 0.15406 nm with a resolution limit of about 12 arc sec. The cross-sectional transmission electron microscope (TEM) images were taken by FEI Tecnai G2 F20 S-TWIN working at 200 kV.

3. Results and discussion

The mosaic structure of the AlGaN epilayer is evaluated by the size and the angular distribution of the mosaic blocks by Williamson–Hall (W-H) plots.[13] Figure 2(a) shows the corresponding W-H plots of (0002), (0004), (0006) reflection planes for triple-axis ω scan of the samples. The straight lines are linear fits of the experimental data, where the $\mathrm{FWHM}_{\omega}\times\sin(\theta)/\lambda$ is given with reciprocal lattice units, θ is the Bragg reflection angle and λ is the x-ray wavelength. From the y-intersection (y_0), the lateral correlation length $L_{||}$ is estimated to be 0.452 μm using $L_{||}=0.9/2y_0$. Since the broadening in symmetric diffractions using W-H plot is only sensitive to the pure screw-type dislocations or the screw-type component of mixed-type dislocations,[4] the determined lateral coherence length $L_{||}$ characterizes the mosaic structure correlated with the screw-type dislocations. The tilt angle α_{Ω} of 0.060 degree is obtained from the slope of the linear dependence. The screw-type dislocations density of 1.0×10^8 cm^{-2} was obtained using Eq. (1).[26]

$$N_{\mathrm{screw}}=\frac{a_{\Omega}^2}{4.35\times b_{\mathrm{scew}}^2}, \tag{1}$$

where the Burgers vector $b_{\mathrm{screw}}=5.074$ Å.

For determination of the heterogeneous strain and vertical correlation length, $\mathrm{FWHM}_{\omega-2\theta}\times\cos(\theta)/\lambda$ is plotted against $\sin(\theta)/\lambda$ for each reflection and again fitted by a straight line as shown in Fig. 2(b). From the y-intersection y_0, the vertical correlation length $L_{\perp}$ (0.767 μm) can be obtained using $L_{\perp}=0.9/(2y_0)$, which is close to the thickness of AlGaN film. The strain value $\varepsilon_{\perp}$ is calculated to be 6.5×10^{-5} from these symmetric reflections.

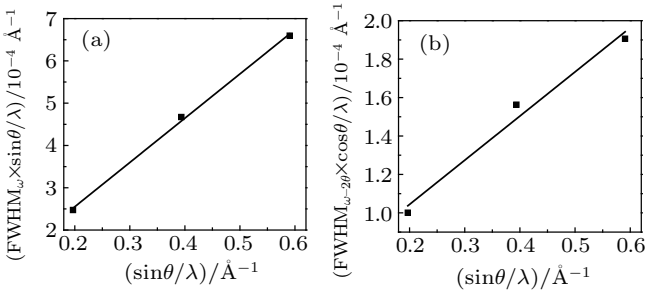


Fig. 2. (a) Triple-axis ω scan for measuring tilt angle and lateral coherence length from W-H plot for AlGaN layers; (b) Triple-axis ω–2θ scan for measuring heterogeneous strain and vertical coherence length of AlGaN layer from W-H plot for AlGaN layers.

The reciprocal space map of $Al_{0.55}Ga_{0.45}N$ $(10\bar{1}5)$ diffraction can be also used to calculate the lateral correlation length and tilt. Figure 3 shows the reciprocal space map taken from $(10\bar{1}5)$ asymmetric reflection, where Q_x and Q_y are the reciprocal space coordinates, which are parallel and perpendicular to the surface normal, respectively.

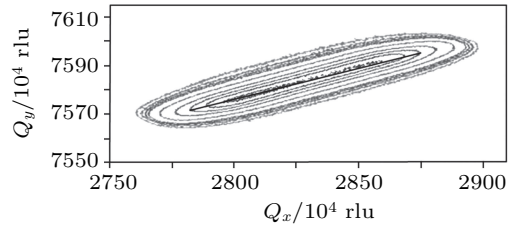


Fig. 3. Reciprocal space map of $Al_{0.55}Ga_{0.45}N(10\bar{1}5)$. The straight line is used for measuring the FWHMs of the overall ellipse.

Using the trigonometry relationships, the lateral correlation length $L_{||}$ can be obtained by Eqs. (2)–(4)

$$\frac{L_2}{L_1}=-\frac{\cos\xi}{\cos(\varphi+\xi)}, \tag{2}$$

$$\frac{L_3}{L_2}=\frac{\sin\varphi}{\cos\xi}, \tag{3}$$

$$L_{||}=\frac{1}{L_1}=-\frac{\sin\varphi}{L_3\cos(\varphi+\xi)}, \tag{4}$$

where $\varphi = \tan^{-1}(Q_x/Q_y)$, $\xi = \tan^{-1}(\Delta Q_x/\Delta Q_y)$,

$$L_3 = \sqrt{(\Delta Q_x)^2 + (\Delta Q_y)^2},$$

and the microscopic tilt is related to the dimension L_2 by Eq. (5)

$$\text{microscopic tilt} = \frac{L_2}{\sqrt{(\Delta Q_x^2 + \Delta Q_y^2)}}, \tag{5}$$

where, L_1 is lateral finite size, parallel to the surface plane; L_2 is the microscopic tilt, normal to the radial direction; L_3 is the overall ellipse, which reflects the FWHM intensity, ΔQ_x and ΔQ_y characterize the full-width-at-half-maximum (FWHM) intensity of the overall ellipse in reciprocal space.[11,20]

The lateral coherence length ($L_{\parallel}$) of AlGaN epilayer was calculated to be 0.048 μm from the RSM measurements, while the tilt angle (0.464 degree) obtained by RSM is much larger than the result (0.060 degree) obtained by W-H plot previously described. Since the orientation of the RSM is sensitive to tilt and lateral coherence length using a dynamic diffraction model developed by Holy *et al.*,[10] an extraction of both $L_{\parallel}$ and α^{tilt} from RSM of $(10\bar{1}5)$ reflection is not feasible. The results indicated that the tilt angle obtained by W-H plots and the $L_{\parallel}$ calculated from RSM are close to TEM observation.

As to determining the twist angle in the mosaic structure of $Al_{0.55}Ga_{0.45}N$ film, the twist angle is measured in off-axis skew symmetric rocking curves of $(10\bar{1}5)$, $(10\bar{1}3)$, $(10\bar{1}2)$, $(10\bar{1}1)$, $(30\bar{3}2)$, and (0002) symmetric planes. The theory of rigid body rotations can be taken into account for the fitting by Eqs. (6) and (7).[14]

$$\alpha^{\text{tilt}}[\varphi] = \cos^{-1}[\cos^2(\varphi)\cos(\alpha_\Omega) + \sin^2(\varphi)], \tag{6}$$

$$\alpha^{\text{twist}}[\varphi] = \cos^{-1}[\sin^2(\varphi)\cos(\alpha_\varphi) + \cos^2(\varphi)], \tag{7}$$

where α_Ω and α_φ are the tilt and twist angles, respectively.

As is shown in Fig. 4, the fitting of data points is utilized to extrapolate FWHMs of asymmetric planes versus inclination angles, and the twist can be extracted by an extrapolation to $\varphi = 90^\circ$. In this case, the result of the twist angle is about 0.392 degree.

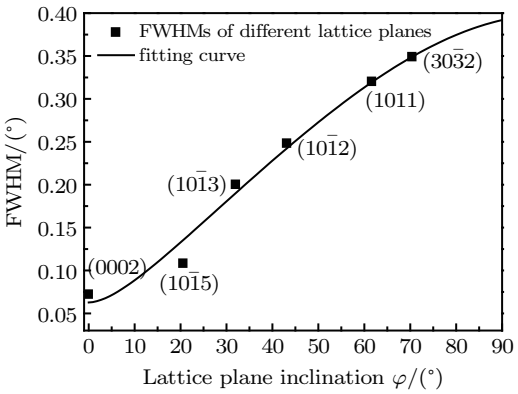


Fig. 4. Measured FWHMs of off-axis skew-symmetric (except (0002) plane) plotted against surface-to-plane angle.

Accordingly, the dislocation density of AlGaN film was calculated from the following Eq. (8).[26]

$$N_{\text{edge}} = \frac{a_\varphi^2}{4.35 \times b_{\text{edge}}^2}, \tag{8}$$

where the Burgers vector $b_{\text{edge}} = 3.147$ Å.

If dislocations are piled up in small grain boundaries to release strain, the density of edge-type dislocations can be obtained by Eq. (9).[27]

$$N_{\text{edge}} = \frac{a_\varphi}{2.1 \times b_{\text{edge}} \times L_{\parallel}}. \tag{9}$$

Thus the density of edge-type dislocation of $Al_{0.55}Ga_{0.45}N$ is 1.8×10^{10} cm^{-2}. The above calculated results are given in Table 1.

Table 1. Summary of the structural parameters of the $Al_{0.55}Ga_{0.45}N$ film obtained by HRXRD and TEM.

Parameter	Value	Method
Lateral correlation length $L_{\parallel}$/μm	0.452	W-H plot
	0.048	RSM
	0.407	TEM (average distance of screw-typed dislocation)
	0.052	TEM (average distance of edge-typed dislocation)
Vertical correlation length $L_{\perp}$/μm	0.767	W-H plot
	0.502	TEM
Tilt angle α_Ω/(°)	0.060	W-H plot
	0.464	RSM
Twist angle α_φ/(°)	0.392	Srikant model[14]
Heterogeneous strain parallel $\varepsilon_{\perp}$	6.5×10^{-5}	W-H plot
Screw dislocation density N_S/cm^{-2}	1.0×10^{8}	W-H plot
	2.0×10^{8}	TEM
	6.0×10^{9}	RSM
Edge dislocation density N_E/cm^{-2}	1.3×10^{10}	α_φ [Eq. (8)]
	1.8×10^{10}	α_φ [Eq. (9)]
	2.8×10^{10}	TEM

TEM analysis is a direct method to characterize defect properties of thin film. Figure 5 shows the cross-sectional TEM images of the sample. The screw-type and edge-type dislocations can be observed under Burgers vectors [0002] and $[11\bar{2}0]$, respectively. Based on counts of threading dislocations, the screw-type and edge-type dislocation densities of the sample are 2.0×10^{8} cm^{-2} and 2.8×10^{10} cm^{-2}, respectively. From cross-sectional TEM images, the lateral correlation length caused by edge-type dislocation is 0.052 μm, and the average distance between screw-type dislocations is 0.407 μm for $Al_{0.55}Ga_{0.45}N$ film, which agrees with the XRD results.

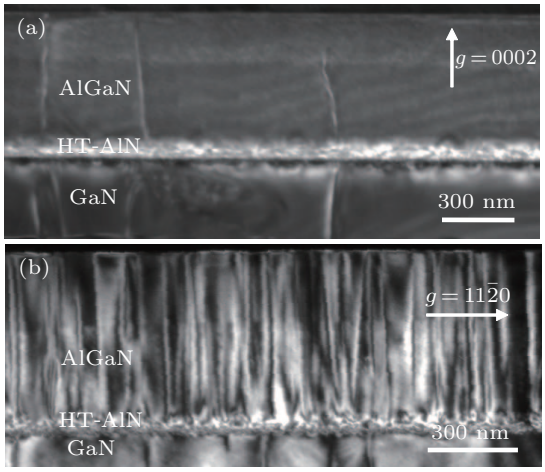


Fig. 5. Cross-sectional TEM images of $Al_{0.55}Ga_{0.45}N$ layer: (a) burger vector of [0002]; (b) burger vector of $[11\bar{2}0]$.

4. Conclusions

The mosaic characteristics of $Al_{0.55}Ga_{0.45}N$ epitaxial layer have been studied by XRD analysis and TEM observation. The structural features including heterogeneous strain, correlation lengths, tilt, twist, and dislocation densities have been determined by means of Williamson–Hall plots and the theory of rigid body rotations from HRXRD. The RSM is also performed for investigation of the AlGaN epilayer to estimate tilt and lateral correlation lengths. The result shows that the lateral correlation lengths obtained by W-H plot and RSM agree with the results obtained by cross-sectional TEM with Burgers vectors [0002] and $[11\bar{2}0]$, respectively. The vertical coherence length of the AlGaN epilayer is consistent with the thickness of AlGaN layer. Furthermore, the screw-type and edge-type dislocation densities obtained from HRXRD are in good agreement with the results observed from TEM, which proves accuracy of evaluation for the dislocation density using the mosaic model.

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