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Short Note

Defect Reduction in $Al_xGa_{1-x}N$ Films Grown by Metal Organic Chemical Vapor Deposition

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The grain size of $Al_xGa_{1-x}N$ films grown on c-plane sapphire substrates was been measured using the X-ray diffraction (XRD) technique. The grain size increases with increasing Al content. It is evident that films with large grain sizes have less defect scattering, and hence, conductivity increases with the grain size. Apparently, the addition of Al to the epilayer increases conductivity. [DOI: 10.1143/JJAP.42.1231]

KEYWORDS: $Al_xGa_{1-x}N$, X-ray diffraction, dislocation density, conductivity

Most GaN (also InGaN and AlGaN) materials used to data for device fabrication are epitaxially grown on sapphire. Because of the large lattice mismatch (14%) between GaN and sapphire, a thin, highly dislocated region is generated at the layer/substrate interface to relieve the strain, and the structural properties of this interface region have been studied here and through out in great detail.[1)] Moreover, the difference of thermal expansion coefficients and lattice constants between GaN and Al_2O_3, induces crystal defects such as micro cracks, micro pipes and mosaic structures, all of which affect the physical properties of GaN films.[2)] So, the calculation of defect densities is very important. Commonly, on axis reflections of high intensity are measured [for example, (00l) reflections for the (001)-oriented epilayer] and their full-width at half maximum (FWHM) values are taken as a figure of merit for the crystalline perfection which is supported by theoretical models.[3)] In contrast, the estimation of total dislocation densities in (001)-oriented GaN epitaxial layers requires the measurement of (hkl) reflections with h or $k \neq 0$.[4)] Such measurements were recently reported for a series of GaN layers grown by hydride vapor phase epitaxy (HVPE)[5)] and metalorganic chemical vapor deposition (MOCVD).[6)] However, systematic studies of the relationship between (00l) and (hkl) X-ray diffraction (XRD) profiles can hardly be found. In this study, we report the reduction of the defects in $Al_xGa_{1-x}N$ films by increasing Al content. The grain size of $Al_xGa_{1-x}N$ films was calculated by using the XRD technique.

The $Al_xGa_{1-x}N$ films were grown by MOCVD on sapphire (0001) substrates with 20-nm low-temperature GaN buffer layers. Trimethygallium (TMGa) and trimethylaluminum (TMAl) were used as metalorganic sources. The nitrogen source used was ammonia (NH_3). The Al content (x) of $Al_xGa_{1-x}N$ films was controlled by the TMAl and TMGa flow rates, and was determined from X-ray diffraction (XRD) spectra by using Vegard's law.[7)] The amount of Al in the $Al_xGa_{1-x}N$ films is linearly dependent on the TMAl/TMGa ratio.

The grain size of the crystal, L, can be estimated by the relation given by Scherrer.[8)]

$$L = \frac{K\lambda}{\Delta\theta_{2\theta}\cos\theta}, \tag{1}$$

where λ is the wavelength of the incident X-rays and K is 0.7[9,10)] for micro crystals and 0.89[11)] for thin films. $\Delta\theta_{2\theta}$ is FWHM of Bragg's angle, and can be calculated by using the following equation.

$$\Delta\theta_{2\theta}^2 = \Delta\theta_{\mathrm{M}}^2 - \Delta\theta_{\mathrm{s}}^2, \tag{2}$$

where $\Delta\theta_{\mathrm{M}}$ is the FWHM of the thin film sample and $\Delta\theta_{\mathrm{s}}$ is the FWHM of the standard sample. We used Al_2O_3 (0001) as the standard sample for growing $Al_xGa_{1-x}N$.

We measured the XRD θ-rocking curve for the (0002) direction (not shown here). The FWHM ranged from 7.2 to 10.8 arcmin with increasing Al content. Figure 1(a) shows the XRD 2θ scan spectrum of the Al_2O_3 substrate. The two peaks corresponding to $Cu_{K\alpha1}$ and $Cu_{K\alpha2}$ of the X-ray source were observed and well resolved by Gaussian fitting. In Fig. 1(a), closed squares denote the measured data, and the dashed and dotted lines show the newly reconstructed peaks. The FWHM of Al_2O_3 is 0.04 due to $Cu_{K\alpha1}$. By the same fitting method, the $Al_xGa_{1-x}N$ samples were fitted and the FWHM values were calculated as 0.11 for GaN [Fig. 1(b)], 0.098 for $Al_{0.03}Ga_{0.97}N$ [Fig. 1(c)], 0.094 for $Al_{0.06}Ga_{0.94}N$ (not shown) and 0.08 for $Al_{0.13}Ga_{0.87}N$ [Fig. 1(d)]. Wang *et al.*[12)] have reported that the surface morphology from SEM observations corresponds to that from the XRD analysis for the (0002) plane. The grain size was calculated and the values ranged from about 800–1000 Å, Al content increasing.

The grain is related to defects such as threading dislocations. As usual, the GaN epitaxial films have a specific defect structure consisting of dislocation ensembles and so-called columns. It has been noted that grain boundaries between grains, as in mosaic structural material, arise only if arrays of dislocations are formed. We deduced the dislocation density from the grain size and the values ranged from 1.4×10^{10} for GaN to $9.9 \times 10^9/\mathrm{cm}^2$ for $Al_{0.13}Ga_{0.87}N$. The dislocation density decreases with increasing Al content. The Ga vacancies could be totally replaced by the diffused Al, even though only one-sixth of the Al can replace the same group-III Ga atoms. It is noted that for the epitaxy methods such as MOCVD and HVPE, rather high temperatures (higher than 800°C) are required for growth. At such sustained elevated temperatures ($T = 1060$°C for MOCVD), atomic diffusion appears to be a dominant process associated

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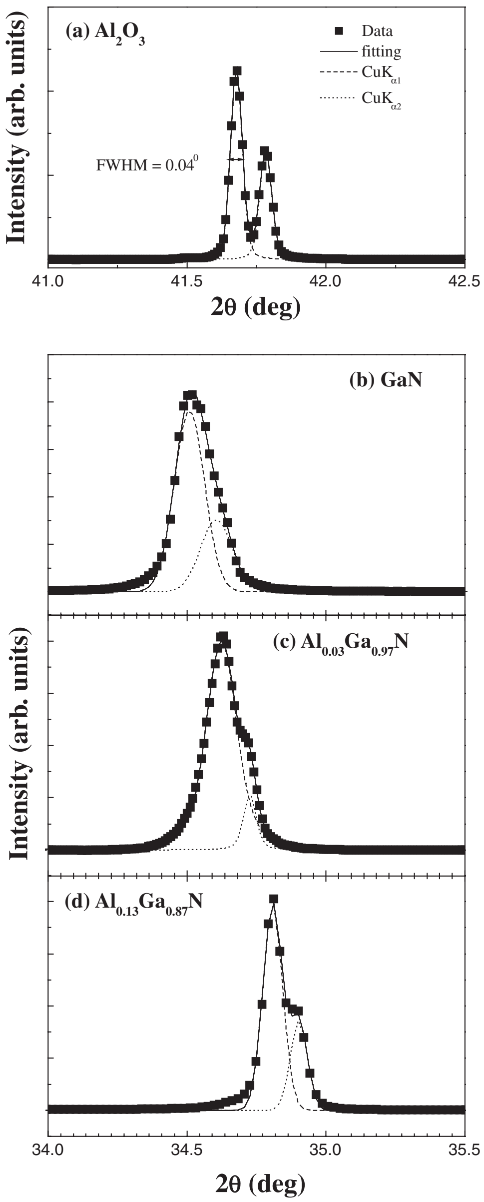

Fig. 1. XRD profiles in the $\theta/2\theta$ mode of $Al_xGa_{1-x}N$ films: (a) Al_2O_3, (b) GaN, (c) $Al_{0.03}Ga_{0.97}N$ and (d) $Al_{0.13}Ga_{0.87}N$. The closed squares represent the data and dotted and dashed lines represent reconstructed data.

with the film's growth, allowing Al atoms to diffuse and fill the Ga sites in the near-interface region in the film. Also, Xu *et al.*[13] have reported that most Ga vacancy sites are replaced by the group-III element Al at the interface. This finding agrees with not only the fact that a copious amount of Al is present at the interface, but also the fact that Al has chemistry similar to that of Ga.

To quantify the scattering effect of dislocations, the inverse of the relaxation time equation is used.[14] In the case of GaN, the dislocation density is $1.4 \times 10^{10}/cm^2$ when obtained by XRD and $7.5 \times 10^9/cm^2$ by the relaxation time equation. Figure 2 shows the relation between conductivity and grain size as a function of Al content. The room-temperature conductivity increases from 5.556 to 66.667/ $\Omega\cdot$cm in accordance with to increasing Al content. This clearly indicates that the conductivity increases with increasing grain size. Given the fact that grain boundaries

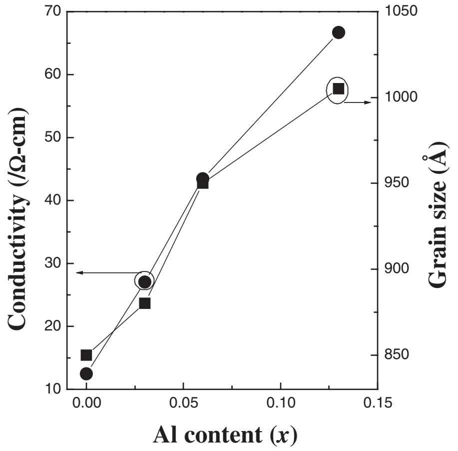

Fig. 2. Conductivity and grain size of $Al_xGa_{1-x}N$ films as a function Al content. The closed circles and squares represent conductivity and grain size, respectively.

are formed by ensembles of dislocations or point defects, the electrons will be scattered by trapped electrons[14,15] when they pass through the boundaries. It is evident that films with large grain sizes have less defect scattering, and hence, the conductivity increases with the grain size. Apparently, the addition of Al to the epilayer increases conductivity. As suggested above, a large grain size (or decreased dislocation density) can be obtained via the increase of Al content.

In conclusion, we determined the grain size of $Al_xGa_{1-x}N$ films from XRD measurement. The grain size of the $Al_xGa_{1-x}N$ films increases with increasing Al content. That is, the grain size ranged from about 800–1000 Å increasing with increasing Al content. The conductivity of the $Al_xGa_{1-x}N$ epilayer increased with increasing grain size.

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