Effects of V/III ratio on species diffusion anisotropy in the MOCVD growth of non-polar *a*-plane GaN films*

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Non-polar *a*-plane ($11\bar{2}0$) GaN films have been grown on *r*-plane ($1\bar{1}02$) sapphire substrates by metal organic chemical vapour deposition. The influences of V/III ratio on the species diffusion anisotropy of *a*-plane GaN films were investigated by scanning electron microscopy, cathodoluminescence and high-resolution x-ray diffraction measurements. The anisotropy of *a*-plane GaN films may result from the different migration length of adatoms along two in-plane directions. V/III ratio has an effect on the growth rates of different facets and crystal quality. The stripe feature morphology was obviously observed in the film with a high V/III ratio because of the slow growth rate along the [$1\bar{1}00$] direction. When the V/III ratio increased from 1000 to 6000, the in-plane crystal quality anisotropy was decreased due to the weakened predominance in migration length of gallium adatoms.

Keywords: non-polar GaN, V/III ratio, anisotropy, migration length
PACC: 8100, 8115H, 7200, 7280E

1. Introduction

Non-polar ($11\bar{2}0$) *a*-plane GaN grown on ($1\bar{1}02$) *r*-plane sapphire substrates has attracted much attention for applications in light emitting diodes because of eliminating the undesirable effects of polarization-induced electrostatic fields along the *c*-axis. Superior properties in comparison with conventional *c*-plane structures have already been reported.[1−3] However, the lattice and thermal expansion mismatches in the non-polar plane are anisotropic. A main difficulty in the non-polar *a*-plane GaN growth lies in the planar anisotropic nature of the growth surface.[4,5] The anisotropic property needs to be further studied in order to improve the crystal quality and explore potential applications for non-polar GaN. The anisotropy of *a*-plane GaN films may result from the different migration length of adatoms along the two directions. It is well known that the V/III ratio has an influence on the growth rates of different planes and could modulate the in-plane anisotropy.[6,7] However, little research related to the V/III ratio influence on the anisotropy in the *a*-plane GaN has been reported so far.

In this work, a series of non-polar *a*-plane GaN films were grown under various V/III ratios on *r*-plane sapphire by MOCVD, and the influences of V/III ratio on the species diffusion in different growth directions of *a*-plane GaN films were investigated by scanning electron microscopy (SEM), cathodoluminescence (CL), and high-resolution x-ray diffraction (HRXRD).

2. Experiment

The growth experiments were carried out in the low-pressure metal organic chemical vapour deposition (MOCVD) system. A 30 nm GaN grown at 600 °C was used as a buffer layer for all the samples. The V/III ratio for the buffer layers was set to be 5000. A 1.5 μm *a*-plane GaN was grown on the GaN buffer layer subsequently. The V/III ratio was varied from 1000 to 6000 for the growth of four kinds of samples. The growth temperature and pressure were fixed to be 1100 °C and 6.67×10^3 Pa. Trimethylgallium (TMG) and NH_3 were used as Ga and N precursors, respectively. Hydrogen was used as a carrier gas. The SEM and CL measurements were conducted

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using MonoCL3 of Gatan which was installed on a JEOL JSM-6700F instrument. The crystal qualities were investigated by HRXRD using a Philips X'Pert MRD system.

3. Results and discussion

Growth anisotropy occurs because the diffusing molecular species are preferentially incorporated into different crystallographic sites. Asai explained trends in the in-plane growth rate anisotropy from GaAs based on the atomic arrangement.[8] A simple model for the lateral growth mechanism is proposed from the consideration of atomic arrangements and the number of dangling bonds at [110] and [$\bar{1}$10] step sites. According to the model, the lateral growth rate is proportional to the number of bonds available for binding Ga atoms at step sites dependent on the growth conditions. Then, the number of dangling bonds and the Ga adatoms should be the key factors for the growth process. In the case of *a*-plane GaN film, the densities of dangling bonds (DB) are different on the (0001), (1$\bar{1}$00) and (11$\bar{2}$0) facets. The difference in the number of nitrogen dangling bonds at the step edge for the in-plane along two-crystal directions causes a difference in the migration length of gallium adatoms. The densities of dangling bonds of (0001), (1$\bar{1}$00) and (11$\bar{2}$0) facets are 11.4, 12.1 and 14 nm^{-2}, respectively.[9] Thus, the growth rate along the [0001] direction should be faster than that of [1$\bar{1}$00], which may result in stripe feature morphology in the growth plane reported in non-polar GaN growth.[10] The growth rate should be also related to the local species density which is strongly dependent on V/III ratios. We further investigated the species diffusion affected by V/III ratios on different growth facets.

Figure 1 shows surface SEM images of the *a*-plane GaN films. There exist different coalescence degrees in these films along the [1$\bar{1}$00] direction and continuous flat surface along the [0001] direction, which confirms the above discussion about the different growth rates due to the different densities of dangling bonds. The film at V/III=1000 in Fig. 1(a) has an almost coalescent surface except for some pits. The films at V/III=2000 and 3000 in Figs. 1(b) and 1(c) have quite rough surfaces with a ridge-like feature along the [0001] direction. Moreover, the film at V/III=6000 has an obvious groove and terrace stripe surface along the [1$\bar{1}$00] direction due to the almost non-coalescence. The different surface morphologies of these films result from the influence of V/III ratio on the growth rates of different planes.

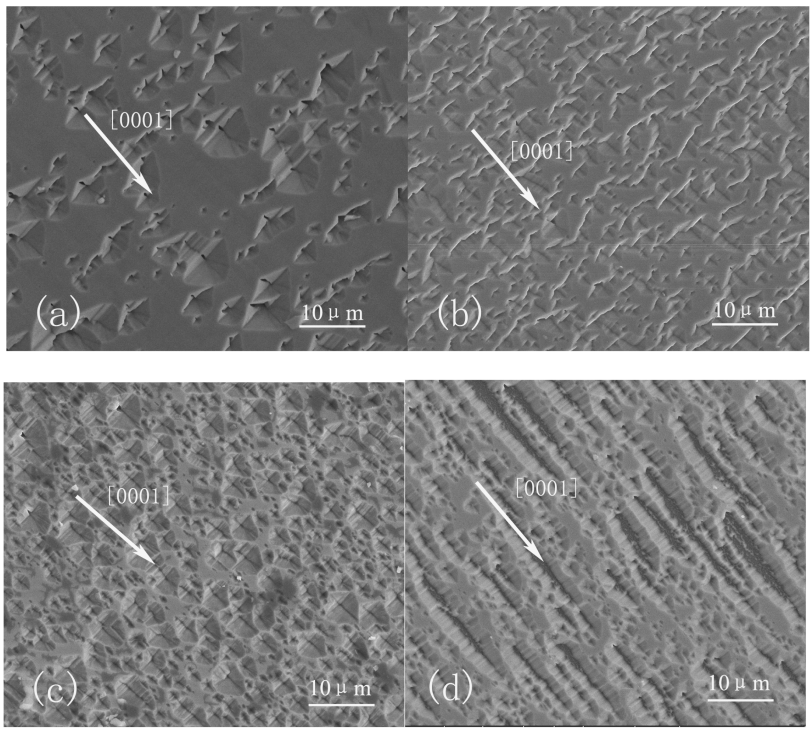


Fig. 1. Surface SEM images of the *a*-plane GaN films: (a) V/III=1000; (b) V/III=2000; (c) V/III=3000; (d) V/III=6000.

Figure 2 gives the growth rates of the vertical [11$\bar{2}$0] direction and the lateral [1$\bar{1}$00] direction as a function of V/III ratios. With increasing V/III ratios, the growth rate decreases in these two directions due to the decreased TMGa partial pressure. Moreover, as shown in the inset in Fig. 2, the growth rate ratio between the [11$\bar{2}$0] versus the [1$\bar{1}$00] direction increases with the V/III ratio from 1000 to 6000. So the stripe feature is most obvious in the film at V/III=6000 due to the slowest [1$\bar{1}$00] lateral growth rate compared with that of the [11$\bar{2}$0] vertical direction.

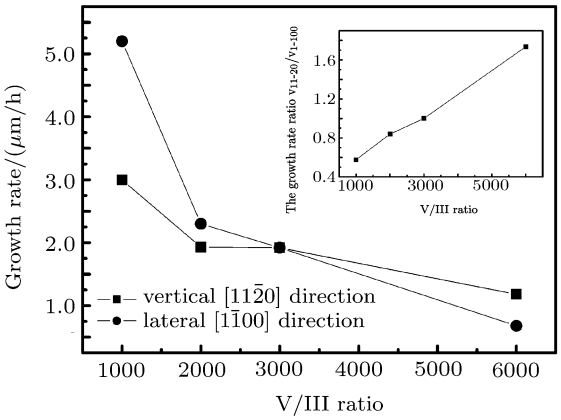


Fig. 2. The growth rates of the vertical [11$\bar{2}$0] direction and the lateral [1$\bar{1}$00] direction as a function of V/III ratios, the inset is the growth rate ratio between the [11$\bar{2}$0] versus the [1$\bar{1}$00] direction.

Other than the dangling bonds on facets, the growth rate is also related to the local species density in the gas phase. The densities of Ga and N bonds are similar on (11$\bar{2}$0) and (1$\bar{1}$00). So the difference of growth rates mainly results from the local species density. The V/III ratio has an influence on the density of Ga atoms on the surface and lateral facet. The Ga atoms preferentially incorporate into the surface and then migrate to the lateral facet. In high V/III ratios, the density of Ga atoms is low, the reaction mainly occurs on the surface. In low V/III ratios with Ga-rich conditions, enough Ga atoms migrate from the surface to the lateral sidewall region. Thereby the density of Ga species for lateral growth increases, and results in a high lateral growth rate at low V/III ratio shown in the inset of Fig. 2.

Figures 3(a)–(d) give the stripe surface CL images of the film at V/III=6000. Figure 3(a) is a panchromatic image and Fig. 3(b)–(d) are monochromatic CL images taken at wavelengths of 370 nm, 330 nm, 575 nm, respectively. The 370 nm luminescence region is the terrace region from GaN. The 330 nm luminescence region corresponding to the strong luminescence in the panchromatic image is from the sapphire substrate, which means that the [1$\bar{1}$00] direction growth rate is very slow under high V/III ratio and there is no coalescence GaN between the terraces. The 575 nm luminescence is almost not observed in the *a*-plane GaN films.

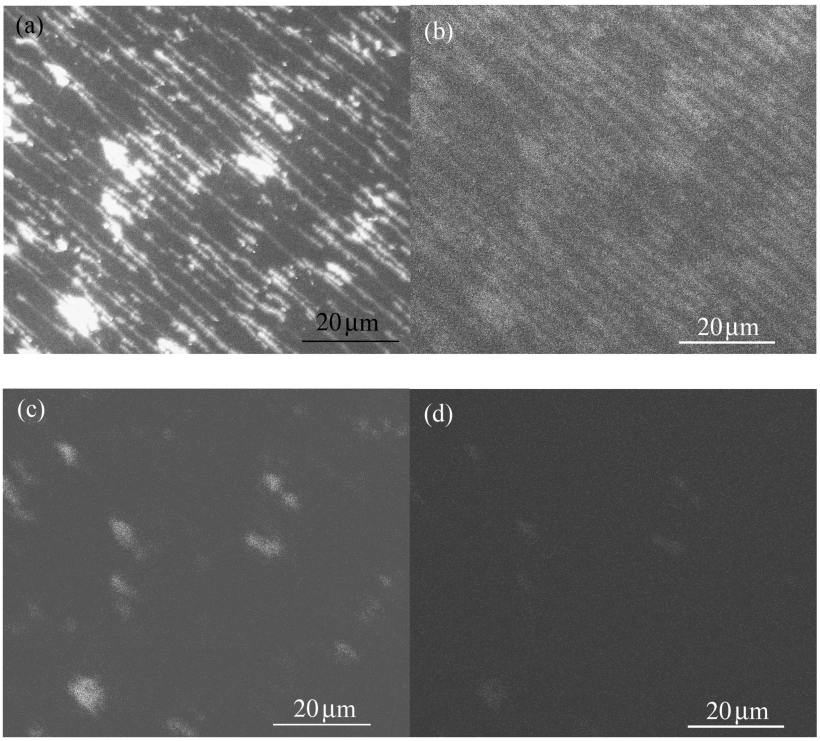


Fig. 3. Stripe surface CL images of the film at V/III=6000: (a) panchromatic; (b) $\lambda = 370$ nm; (c) $\lambda = 330$ nm; (d)$\lambda = 575$ nm.

The results further indicate that the growth rate in the $[1\bar{1}00]$ direction at high V/III is too small to coalesce the GaN terrace and sapphire is exposed in the groove regions. The effect of V/III ratios on the growth rates of different planes results in the different surface morphologies.

The species diffusion anisotropy should give rise to a variation of crystal quality of different planes. The anisotropic crystal qualities in the growth plane can be evaluated by the full width at half maximum (FWHM) of x-ray rocking curves as a function of azimuth angle in HRXRD measurements. Figure 4 shows its evolution with V/III ratio. Azimuth angles of 0 and 90 separately correspond to the projection of the incident x-ray beam parallel to the [0001] and $[1\bar{1}00]$ directions, respectively. It is clear that the FWHM is largest along the $[1\bar{1}00]$ direction and is smallest along the [0001] direction, which corresponds to the better crystal quality in the [0001] direction than the $[1\bar{1}00]$ direction. This anisotropy could come from the atomic arrangements on the $(11\bar{2}0)$ a-plane GaN surface.[11] The difference in the number of nitrogen dangling bonds at the step edge for the two directions [0001] and $[1\bar{1}00]$ causes a difference in the migration length of gallium adatoms. The gallium adatoms can migrate a long distance at the step edge with a low density of nitrogen dangling bonds, so that the migration length in the [0001] direction is much longer than that in the $[1\bar{1}00]$ direction. The film quality is better in the [0001] direction with longer migration length. With increasing V/III from 1000 to 6000, such anisotropy of crystal quality is decreased. In the sample of V/III=6000, the FWHMs of different azimuths are almost the same. With increasing V/III from 1000 to 6000, the density of Ga adatoms is decreased, and the predominance in the direction [0001] is weakened, which results in a reduced quality anisotropy. Therefore, the crystal quality anisotropy could be modulated by optimizing the V/III ratios.

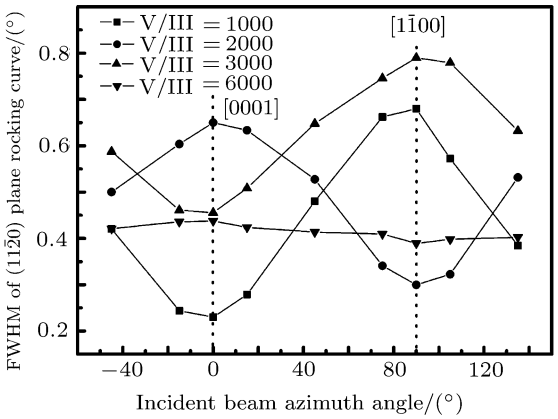


Fig. 4. FWHM of x-ray $(11\bar{2}0)$ omega scans for samples with different V/III ratios as a function of azimuth angles.

4. Conclusions

In summary, we have investigated the effects of V/III ratio on the species diffusion anisotropy of a-plane GaN films. The anisotropy of a-plane GaN films may result from the different migration length of adatoms along two directions in the a-plane. V/III ratios have an influence on the local species densities of Ga atoms on the surface and lateral facets. The stripe feature was obviously observed in the film with high V/III ratio because of the slow growth rate along the $[1\bar{1}00]$ direction. When V/III ratios were increased from 1000 to 6000, the in-plane crystal quality anisotropy was decreased due to the weakened predominance in migration length of gallium adatoms.

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