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Flow-rate modulation epitaxy of wurtzite AlBN

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1. Introduction

Nitride semiconductors containing boron atoms are attractive candidates for light emitting devices in the deep-UV region because of their wide bandgaps and complete lattice-matched epitaxy on various substrates, such as SiC, AlN, or GaN. Wurtzite AlBN (w-AlBN) and wurtzite BGaN thin films have been grown by metalorganic chemical vapor deposition (MOCVD) [1] and molecular beam epitaxy [2]. A large number of critical issues, however, have strongly limited the crystallinity of nitride semiconductors containing boron atoms. These serious problems include phase separation due to the difference in the stable crystal polytypes (graphite-type for BN, but wurtzite for the other nitrides), extremely high growth temperatures, and parasitic reactions in the gas phase. Flow-rate modulation epitaxy (FME) is a useful epitaxial growth method for compound semiconductor thin films, wherein group-III and group-V sources are alternatively supplied to the growing surface.[3] This technique enables us to lower the optimal growth temperatures, because the surface migration of group-III metal atoms are enhanced in the absence of excess surface coverage of group-V atoms. Furthermore, the alternate supply of source molecules can suppress the parasitic reactions in the gas phase. This study applied FME to grow w-$Al_{1-x}B_xN$ thin films on SiC substrates. The results show that FME is more useful for growing w-$Al_{1-x}B_xN$ thin films with smooth surfaces and good crystallinity than conventional MOCVD.

2. Experimental

AlBN films were grown on (0001) Si-oriented n-type 4H-SiC substrates using a vertical-type MOCVD apparatus. Reaction pressure and carrier gases were 300 Torr and a mixture of N_2 and H_2, respectively. AlBN films were directly grown on SiC substrates at 1000 ℃. The thickness of AlBN was 350 or 780 nm. The source gases were triethylboron (TEB), trimethylaluminum (TMA), and NH_3. A 2-sec. supply of metalorganics and a 1-sec. supply of NH_3 were alternately repeated without growth interruption in the FME procedure. AlBN thin films were also prepared by conventional MOCVD (simultaneous supply of metalorganics and NH_3) for comparison. Growth rates were evaluated by *in-situ* monitoring of optical reflectance with a He-Cd laser (325 nm). The surface morphology of the AlBN thin films was observed by atomic force microscopy (AFM). The crystallinity of AlBN was evaluated by ω–2θ scans in high resolution X-ray diffraction (HR-XRD). The boron composition was evaluated by reciprocal lattice maps in HR-XRD. The crystallinity and orientation of the films were evaluated by cross-sectional high resolution transmission microscopy (X-HRTEM).

3. Results and discussion

In Fig. 1, the growth rates of AlBN films are plotted as a function of the flow rates of metalorganics, [TMA]+[TEB]. In these experiments, the flow rate ratio of [TEB]/{[TMA]+[TEB]} was kept constant at 0.04. The growth rates of AlN are also plotted for comparison. The growth rates of AlBN and AlN increase linearly with increasing the flow rates of metalorganics. The proportional increase of the growth rate implies that the parasitic reactions between NH_3 and metalorganics are effectively eliminated. In conventional MOCVD, the parasitic reactions between NH_3 and TMA (TEB) have been a crucial issue. All the FME growth described from the next paragraph was performed by using the growth rate of approximately 1 ML/cycle.

Figure 2 shows evolution of optical reflectance at λ= 325 nm during the growth of 350-nm-thick AlBN films for conventional MOCVD and FME. In both cases, the periodic optical fringes are clearly observed, where one period corresponds to the AlBN thickness of 67 nm. The optical reflectance of FME has both constant amplitude and constant peak values throughout the growth. This result indicates that the AlBN film grows two-dimensionally and the surface remains quite smooth throughout the entire growth in the FME method. It is considered that the elimination of parasitic reactions in gas phase and the enhancement of the surface migration of Al and B atoms result in the two-dimensional and homogeneous growth of an AlBN film by FME. On the other hand, amplitude of the optical fringes for conventional MOCVD is damped gradually, indicating that the surface roughness of the AlBN film increases with increasing its thickness. The mean roughness of the AlBN surfaces measured by AFM was 10.5 and 1.0 nm for MOCVD and FME, respectively. It can be concluded that FME is very useful for growing an AlBN film with a smooth surface, compared with conventional MOCVD.

Figure 3 shows the ω–2θ scan charts for 350-nm-thick AlBN films fabricated by MOCVD and FME. The AlBN film grown by MOCVD exhibits a broad peak at 18.059 degrees accompanied by a very weak peak at 18.947 degrees. The peak at the lower angle is considered to be from w-AlBN(0002) planes. The weak peak at 18.947 degrees is from w-AlBN(1$\underline{1}$01) planes. From X-HRTEM observation, we confirmed that polycrystallization occurs during the growth of AlBN, resulting in the mixing of a small amount of the faceted (1$\underline{1}$01) AlBN grains in the (0001) AlBN matrix. In contrast, the AlBN film obtained by FME shows a single sharp peak at 18.095 degrees without an additional peak up to 22 degrees (The w-BN(0002) peak position is 21.372 degrees), indicating that the w-AlBN film grown by FME was c-axis oriented singlecrystal. Further analysis by reciprocal lattice maps in HR-XRD revealed that the boron composition (x) in the $Al_{1-x}B_xN$ film grown by FME is 0.015.

4. Conclusions

FME is useful for improving the quality of w-AlBN thin films. A 350-nm-thick w-$Al_{0.985}B_{0.015}N$ film grown by FME exhibited a single sharp peak in an ω–2θ scan of HR-XRD and a smooth surface with mean roughness of 1.0 nm. In contrast, w-AlBN films grown by conventional MOCVD contained nano-sized w-AlBN(1$\underline{1}$01) crystallites in the w-AlBN(0001) matrix. FME is also effective in suppressing this polycrystallization because it enhances the surface migration of B atoms.

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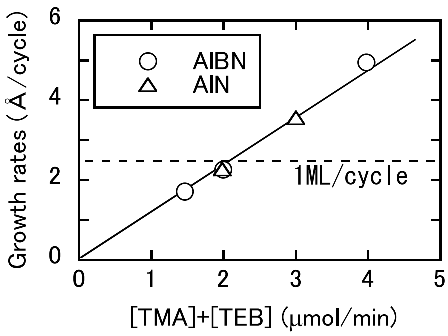

Fig. 1: Growth rate of AlBN thin films (open circles) as a function of the flow rates of metalorganics, [TMA]+[TEB], for FME growth. The growth rates of AlN (open triangles) are also plotted for comparison.

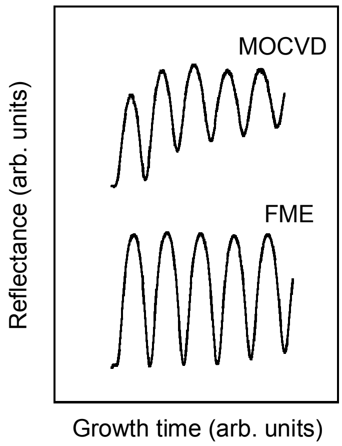

Fig. 2: Evolution of optical reflectance at λ= 325 nm during the growth of 350-nm-thick AlBN films for conventional MOCVD and FME, respectively.

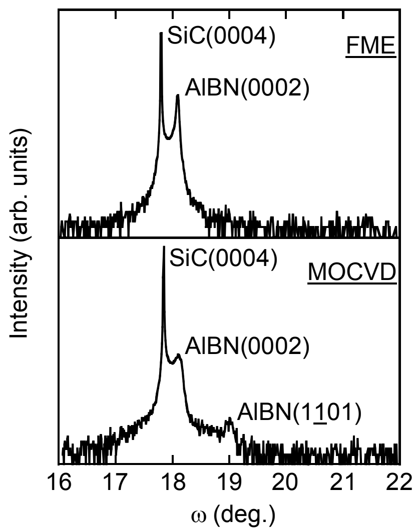

Fig. 3: The ω–2θ scan charts for 350-nm-thick AlBN films fabricated by MOCVD and FME.