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Characterisation of GaNP Layers Grown on (0001) GaN/Sapphire by Gas Source Molecular Beam Epitaxy

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1. Introduction

Much attention has been paid to the study of Ⅲ-Ⅴ nitride-related materials because of their application in short wavelength optoelectronics and high temperature electronics. Blue-green light emitting diodes and violet laser diodes, which can be operated at room temperature, was already demonstrated [1]. This material system can be applied to a wide range of wavelengths from ultraviolet to larger than 2 μm by adding As or P, because of the large bowing of band gap energy [2]. A large miscibility gap is expected in GaN-GaP system as a result of the large differences in lattice structure (GaN: wurtzite, GaP: zinc-blende) and in lattice mismatch (~20 %) [3]. This large miscibility gap would hinder the successful growth of this material. Until now, several authors reported the growth of GaPN and GaAsN [4-10]. However, there is a lack of detailed microstructural characterisation of GaNP (0001) layers grown by MBE.

In this work, comprehensive transmission electron microscopy (TEM) and transmission electron diffraction (TED) examination was performed to investigate the phase stability and microstructures of $Ga_xN_{1-x}P$ layers grown on (0001) GaN by gas source MBE at temperatures ranging from 500 °C to 750 °C.

2. Experimental details

The GaNP layers examined were grown on metalorganic vapour phase epitaxial (MOVPE) (0001) GaN/sapphire substrates (from Cree Research Inc.) in a modified GEN Ⅱ GSMBE system. The growth temperatures ranged from 500 °C to 750 °C. A nitrogen flow of 3 SCCM was introduced to the RF source (Oxford Applied Research). PH_3, cracked at 1000 °C, provided the P flux and its flow rate was between 0 and 3 SCCM. The background pressure during growth was in the range 3–5x10^{-5} Torr, and the growth rate was 0.1 - 0.15μm/h which was limited mainly by the nitrogen flow rate. Details of growth conditions for each sample are given in Table I.

For electron microscope examination, [2-1-10] and [01-10] cross-section, and [0001] plan-view thin foil specimens were prepared using standard procedures and finishing by Ar^+ ion thinning with the specimen cooled to ~77K. TEM, TED, and high resolution electron microscopy (HREM) examination was performed in a JEM 2010 instrument operated at 200kV.

Table I. Growth details of GSMBE grown $Ga_xN_{1-x}P$ layers

Sample	Growth Temp. (℃)	PH_3 flow rate (SCCM)	N_2 flow rate (SCCM)	Phases observed
#2184	500	3	3	β
#2183	600	3	3	β
#2186	730	3	3	β
#2182	750	3	3	β

3. Results and Discussion

Figure 1 shows cross-section TED pattern taken from a thin region containing the GaNP layer (grown at 500 °C) and the GaN substrate, with electron beam aligned along the $[01\text{-}10]_{GaN}$ direction. There are well-defined sharp spots from the GaN substrate, and weak diffraction spots which are associated with the rings of diffuse diffracted intensity. The diffuse rings are attributed to the randomly oriented polycrystalline structures of the zinc-blende GaNP layer. The ring (indicated by 'a') corresponds to the {111} spots of GaP-rich side of GaNP. The second (indicated by 'b'), the third (marked 'c'), the forth (marked by 'd'), and the fifth rings (marked 'e') correspond to the {220} spots of GaP-rich side of GaNP, the {220} spots of β-GaN, the {222} spots of β-GaN, and the {024} of GaP, respectively. The TED results show that the binary alloys of β-GaN and GaP, and the ternary alloy GaNP are formed at a low temperature. The TEM dark field images (not shown) exhibited the growth of columnar grains, which was consistent with the TED results.

The TEM and TED results obtained from the GaNP layer grown at 600 °C showed that the growth behaviour was similar to those of the GaNP layer grown at 500 °C. TEM DF results, however, showed that columnar grains were inclined 50° ~ 60° from the $[0001]_{GaN}$ direction.

In Fig. 2(a) is shown cross-section $[2\text{-}1\text{-}10]_{GaN}$ TED pattern obtained from the GaNP layer grown at 750 °C. The pattern is indexed in Fig. 2(b). The TED results show that the GaNP layer has a zinc-blende structure. As can be seen in Fig. 2(b), two cubic [110] TED patterns are superimposed, with the {111} planes parallel to the $[0001]_{GaN}$ direction. Combined with syncrotron x-ray scattering results, TED examination shows that there are two different types of crystal domains, each having twin relation to the other.

In Fig. 3 is shown (002) cross-section DF image obtained from the GaNP layer grown at 750 °C. The image reveals that there are fine needle-like contrast (termed here

'micro-domains') lying nearly parallel to the GaN/GaNP interface. The micro-domains vary in width from ~ 0.8 nm to ~ 2.5 nm and in length from ~ 2 nm to ~ 10 nm. The (001) plan-view TEM DF results showed that the domains were fine rod-shape. The presence of these rod-like micro-domains seems to be responsible for the occurrence of the diffuse diffracted streaks along the [0001] growth direction in Fig. 2(a).

4. Summary

TEM and TED examination was made of GSMBE GaNP layers grown on (0001) GaN/sapphire substrates grown at temperatures in the range 500 – 750 °C to investigate microstructures and phase stability. TED and TEM results show that the GaNP layers grown at T<600 °C are polycrystalline with a zinc-blende structure. As for the 500 °C layer, the grains are randomly oriented. For the 600 °C layer, however, the grains are textured with the specific direction, i.e. inclined 50 – 60° from the $[0001]_{GaN}$ growth direction. The layers consist of a mixed phase of GaNP and GaN. The GaNP layers grown at T > 730 °C are single crystalline with a zinc-blende structure. DF results show that for the layers grown at T>730 °C there exists two different types of micro-domains, each having twin relation to the other. A simple model is suggested to explain how the P flux influences the microstructures of the GaNP layers.

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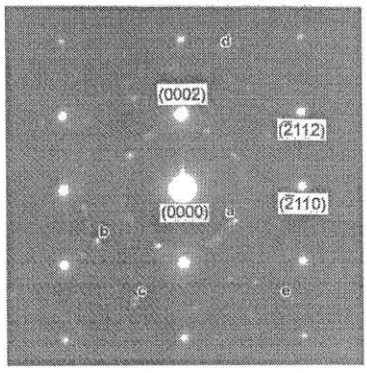

Fig.1 $[01\text{-}10]_{GaN}$ cross-section TED pattern obtained from MBE GaNP layer grown at 500 °C.

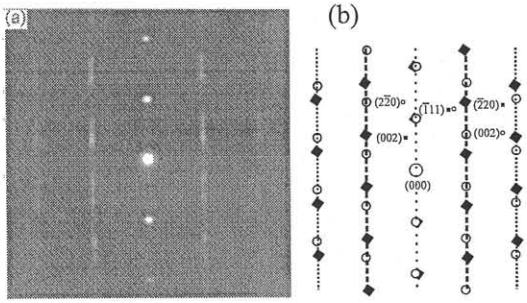

Fig.2 (a) $[2\text{-}1\text{-}10]_{GaN}$ TED obtained from MBE GaNP layer grown at 750 °C, (b) The pattern is indexed.

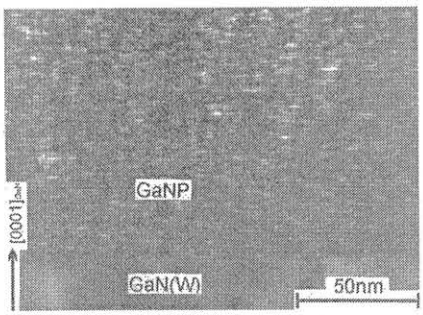

Fig.3 (002) dark field image obtained from MBE GaNP layer grown at 750 °C.