Nitrogen source로 암모니아, DMH_y(dimethylhydrazine)을 사용해 Gas-Source MBE로 성장된 InGaN 박막특성

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Growth of InGaN on sapphire by GSMBE(gas source molecular beam epitaxy) using DMH_y(dimethylhydrazine) as nitrogen source at low temperature

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Abstract

High quality GaN layer and $In_xGa_{1-x}N$ alloy were obtained on (0001)sapphire substrate using ammonia(NH_3) and dimethylhydrazine(DMH_Y) as a nitrogen source by gas source molecular beam epitaxy(GSMBE) respectively. As a result, RHEED is used to investigate the relaxation processes which take place during the growth of GaN and $In_xGa_{1-x}N$. The full Width at half maximum of the x-ray diffraction(FWHM) rocking curve measured from plane of GaN has exhibitted as narrow as 8 arcmin. Photoluminescence measurement of GaN and $In_xGa_{1-x}N$ were investigated at room temperature, where the intensity of the band edge emission is much stronger than that of deep level emission. In content of $In_xGa_{1-x}N$ epitaxial layer according to growth condition was investigated.

Key Words : $In_xGa_{1-x}N$, GSMBE, dimethylhydrazine(DMH_y)

1. Introduction

Since the accomplishment of Nakamura and coworker on GaN based light diode[1] and lasers[2], the Ⅲelement nitrides are attracting tremendous interest as candidate materials for optoelectronics devices in blue and near-UV spectrum regions because GaN and Ⅲelement nitrides has large direct band-gap energy at room temperature. Despite several difficulties in the epitaxial of Ⅲ-nitrides, such as the lack of absence of well-suited substrate, High quality GaN/$In_xGa_{1-x}N$ is obtained elsewhere. Molecular beam epitaxy (MBE) is however rapidly developing. Low temperature used in MBE compared to MOCVD could be determinant for growing $In_xGa_{1-x}N$ alloy with high In content. The recent studies revealed strong dependence of indium incorporation into the InGaN tenary compounds in the number of process parameters, such as growth temperature[3], both indium and gallium incident fluxes[3,4], Ⅲ/V ratio[4]. Generally, Low efficiency and nonuniformity of In incorporation observed in the experiments was attributed to low thermal stability of InN[5], phase separation[6] as well as indium droplet formation on the growth surface[4]. However the role and importance of each of these factors are still poorly understood.

In this paper we report on the use of RHEED tomonitor the growth of $In_xGa_{1-x}N$ layers on GaN. The In mole fraction has been determined from RHEED, XRD, STM and PL. Therefore it is necessary for the proper choice of the process parameters allowing growth of $In_xGa_{1-x}N$ ternary compounds. The incorporation rate of In as *different growth parameter investigated.*.

2. Experiment

GaN layer and InGaN alloy were grown on(0001)sapphire substrate in Gas-source MBE system equipped with standard Knudsen cell for Ga, In and with gas source for NH_3, DMHY supply and with RHEED for in-situ monitoring the growth processand with pryrometer for measuring a substrate temperature. The atomic nitrogen species were provided by the decomposition of NH_3 on the GaN layer and of DMH_Y on the InGaN layer. The decomposition of NH_3 begins from 400℃ to 800℃ at high temperature. The incorporation efficiency is 0.5% at 500℃and become 3.8% at 700℃. Above 700℃, the actual efficiency of cracking is higher since a part of the nitrogen must have been lost by evaporation and did not contribute to the formation of the epilayer[5,6]. However, DMH_Y decomposed at low temperature(below 600℃)but introduces carbon impurity in the epilayer. During the growth, carbon is included in 0.155 compare to 0.0232 at MOCVD. Prior to the experiments, the c-plane sapphire(0001) substrate was cleaned with organic solvent, then etched in boiling aqua regia($3HCl:HNO_3$) for 10min, and finally loaded into a GSMBE system, in which the base pressure was $5*10^{-8}$Torr. The first step of the growth consists in performing a nitridation of the sapphire during 10 min at substrate temperature 900℃ under $5*10^{-5}$Torr of NH_3. This results in the formation of an AlN thin layer[7,8]. Then GaN buffer layer was deposited during 3min at substrate temperature 400℃ ~600℃ under varied V/Ⅲ ratio. The layers of GaN were grown using NH_3 at substrate temperature 850℃ during 120 min under varied V/Ⅲ ratio.

Table 1. condition of InGaN epilayer

substrate annealing	T_S	950℃
	Time	10 minute
Nitridation	T_{Sub}	900℃
	P_{NH3}	$5.00*10^{-5}$Torr
	Time	10 minute
GaN Buffer layer	T_{Sub}	400℃
	P_{Ga}	$2.0*10^{-7}$Torr
	P_{NH3}	$2.5*10^{-5}$Torr
	Thickness	20nm
GaN epilayer	T_{Sub}	850℃
	P_{Ga}	$2.0*10^{-7}$Torr
	P_{NH3}	$5.0*10^{-5}$Torr
	Thickness	200nm
InGaN epilayer	T_{Sub}	600-750℃
	P_{In}	$1-2.0*10^{-7}$Torr
	P_{Ga}	$1-3.0*10^{-7}$Torr
	P_{DMHy}	$1-3.0*10^{-4}$Torr
	Growth time	120 minute

then GaN buffer layer and GaN epitaxy layer were deposited in turn at 400℃ at P_{Ga} : $2.0*10^{-7}$ Torr, P_{NH3} : $2.5*10^{-5}$ Torr, 850℃ at P_{Ga} : $5.0*10^{-7}$Torr, P_{NH3} : $5.0*10^{-5}$Torr. and In content of $In_xGa_{1-x}N$ epitaxial layer according to growth condition was investigated by using DMH_y as nitrogen source at low temperature.

2. Experimental results and discussion

2.1 The growth of GaN with NH_3

GaN buffer layer formed on the sapphire surface after nitridation according to growth condition at P_{Ga}:2.0*10^{-7}Torr, P_{NH3}:5.0*10^{-5}Torr:, P_{Ga}:3.0* 10^{-7} Torr, P_{NH3}:5.0*10^{-5}Torr P_{Ga}: 2.0*10^{-7} Torr, P_{NH3} :2.5*10^{-5}Torr. GaN layer formed on the GaN buffer layer (P_{Ga}:2.0*10^{-7}Torr, P_{NH3}:5.0*10^{-5}Torr) according to P_{Ga}:2.0*10^{-7}Torr, P_{NH3}:5.0*10^{-5}Torr P_{Ga}:3.0*10^{-7}Torr, P_{NH3}:5.0*10^{-5}Torr P_{Ga} : 5.0*10^{-7} Torr P_{NH3} : 5.0*10^{-5}Torr. The standard rocking curve of a GaN layer, with full width at half-maximum of 8 arcmin, is shown in Fig 1. Their root mean square(rms) roughness measured by atomic force microscopy(AFM) on GaN layer varies from 4.57 to 7.83nm when changing the V/III ratio in P_{Ga}:2.0*10^{-7}Torr, P_{NH3}:5.0*10^{-5}Torr P_{Ga}:3.0*10^{-7}Torr, P_{NH3}:5.0*10^{-5} Torr and P_{Ga}:5.0*10^{-7}Torr, P_{NH3}:5.0*10^{-5} Torr.

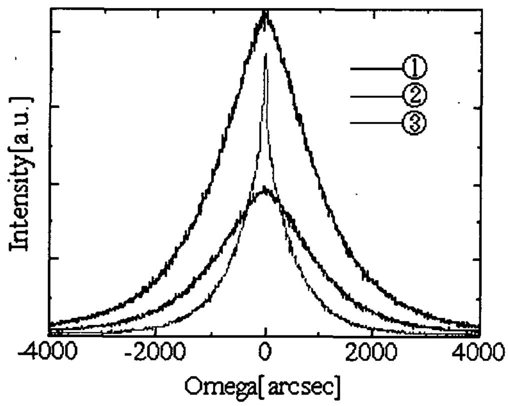

Fig 1. XRD measurements of GaN layers with ①P_{Ga} : 2.0*10^{-7}Torr, P_{NH3} : 5.0*10^{-5}Torr, ②P_{Ga} : 3.0*10^{-7}Torr, P_{NH3} :5.0*10^{-5}Torr, ③P_{Ga} : 5.0*10^{-7}Torr, P_{NH3} : 5.0*10^{-5}Torr

2.2 The growth of InGaN with dimethlyhydrazine

Recently many articles have appeared on the growth of InGaN alloys using both MOVPE and MBE. Yoshida[9]grew InGaN epilayers which In concentration was 20% by MBE on sapphire substrate. He found that the growth temperature had a large effect on the quality of the layers. If the growth temperature was more than 800℃, the concentration of In was very low. On the other hand if the temperature was lower than 760℃, Ga droplets were formed on the surface because of low decomposition rate of ammonia. Grandjean and Massies[6] have grown $In_xGa_{1-x}N$ layers with high In concentrations (up to x= 0.46)by GSMBE. Since at low temperatures the fraction of ammonia that decomposes is smaller, the growth rate was reduced from 1.2 to 0.2-0.4micro/hour. To obtain reactive species of nitrogen at low temperature, we used DMHy as nitrogen source. DMHy decomposes at low temperatures below 600℃ but introduces carbon impurity in the epilayer and we investigated that the quality of InGaN epilayer on the incorporation of In. At first, we investigated InGaN had been grown at the temperature of 650℃, 700℃, 750℃ (P_{Ga} : 1.0*10^{-7}Torr, P_{In} : 1.0*10^{-7}Torr, P_{DMHy} : 1.010-4 Torr) with DMHy as nitrogen source. When growth temperature is 650℃, It appeared In-droplet. At that temperature, the cracking efficiency of DMHy on

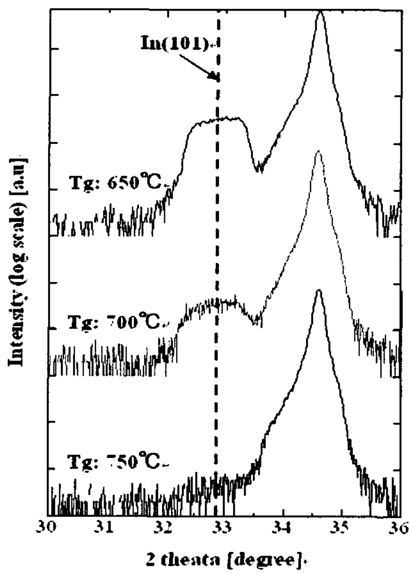

Fig 2. XRD result of InGaN on GaN according to the temperature($2\theta - \omega$ scan)

the GaN surface is low. But according to increasing the growth temperature, In droplet is disappeared. In concentration of InGaN was calculated by Vegard law.

$$3.34eV = 1.9x + 3.4(1-x) - bx(1-x)$$

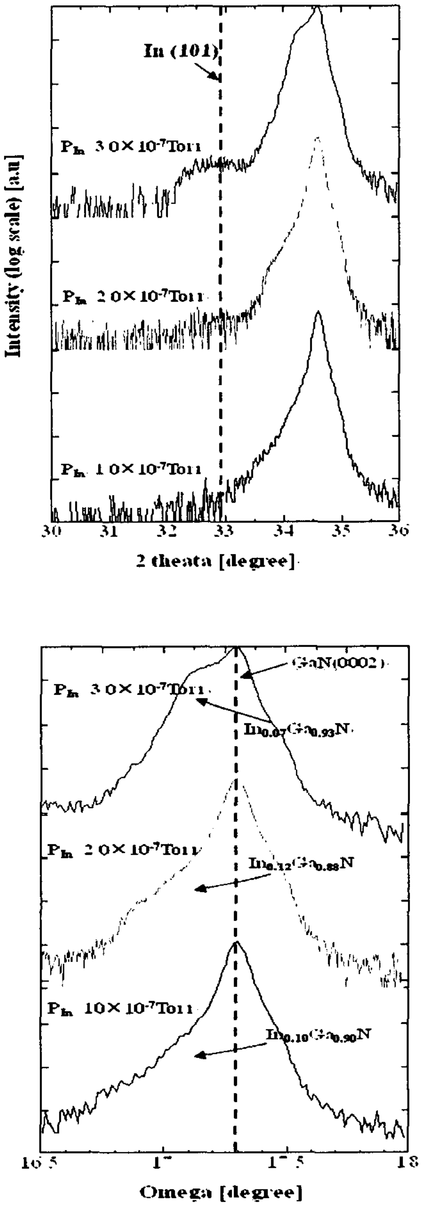

Fig 3. XRD result of InGaN on GaN according to the varied P_{In} $2\theta-\omega$ scan, ω scan

The bowing parameter obtained was 0.95eV, which agreed with that obtained for the samples grown at substrate temperature. At growth temperature of 750℃, In concentration of InGaN obtained 12%. But the morphology of surface was rough (rms = 10.35nm). At second, InGaN had been grown at the In beam equilibrium pressure of P_{In}:1.0*10^{-7}, 2.0*10^{-7}, 3.0*10^{-7} (T_{sub}:650℃, P_{Ga}:1.0*10^{-7}, P_{DMHy} : 2.0*10^{-4}Torr). Figure 3 show according to the In BEP. In concentration of InGaN epilayer is 7%, 12%, 10% by the Vegard law. In concentration of InGaN is not linearly at the increasing in In BEP. The coupling energy of InN is 1.9eV, however GaN is 2.5eV[10]. At that result, InN is more difficult to combine than GaN in the InGaN. The measurements of AFM were 6.33 , 3.56 and 3.11 respectively. In order to determine an optimal condition, we investigated photoluminescence at the room temperature. we measured that GaN in P_{Ga} :2.0*10^{-7}Torr, P_{NH3}: 5.0*10^{-5}Torr at the growth temperature 850℃ (FWHM=8 acrmin)and InGaN on that GaN in P_{In}: 3.0*10^{-7}Torr, P_{Ga} : 1.0*10^{-7}Torr, P_{DMHy} : 2.0*10^{-4}Torr at growth temperature 650℃. The PL of that sample was shown in Figure 4. In the case of GaN, a sharp peak at 365nm in the band to band and a broad peak at 575nm for a deep level were observed. The FWHM at the band-to-band emission was 8 arcmin. The yellow luminescence in GaN is a broad luminescence band centered around 2.2-2.3eV. The YL appears to be a universal feature. It has been observed in bulk GaN crystallites as well as in epitaxial layers grown by techniques as different as molecular beam epitaxy (MBE), metalorganic chemical vapor deposition (MOCVD), and hydride vapor phase epitaxy (HVPE). The deep level introduced by the Ga vacancy or related compound is responsible for the broad peak YL in GaN and the formation of Ga vacancy may be enhanced by complex formation with donor impurities[11].. Therefore, according to the increasing V/Ⅲ ratio yellow luminescence was observed but every samples were appeared YL in the broad peak at 575nm. In the case of InGaN, One peak was a GaN band-to-band emission (365nm) of the GaN epilayer ; the other was a band-to-band emission (371nm) of InGaN.

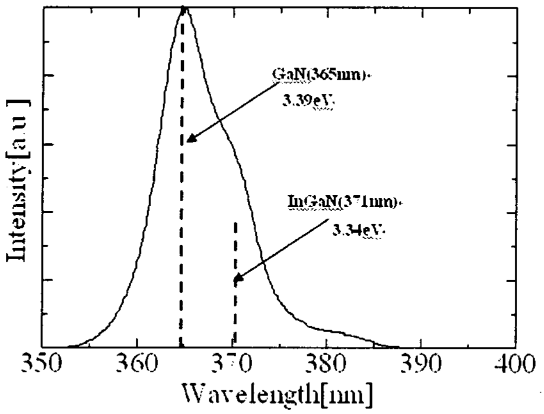

Fig 4. Photoluminescence spectrum radiated InGaN film at room temperature, Excitation laser is He-Cd^{+} laser

4. conclusions

The GaN and $In_xGa_{1-x}N$ were grown using the GSMBE method. GaN and $In_xGa_{1-x}N$ crystal were obtained using ammonia, DMHy as the source of reactive nitrogen. We have demonstrated that the quality GaN was studied according to the varied growth condition and InGaN was grew on GaN using DMHy as nitrogen source. Photoluminescence measurement of $In_xGa_{1-x}N$ were investigated. There band to band luminescence and yellow luminescence were observed. The intensity of the band edge emission is much stronger than that of deep level emission. In content of $In_xGa_{1-x}N$ epitaxial layer according to growth condition was investigated.

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