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Characterization of p-type $In_xGa_{1-x}N$ grown by metalorganic chemical vapor deposition

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Abstract

This study investigates the electrical and optical characteristics of Mg-doped $In_xGa_{1-x}N$ grown by metalorganic chemical vapor deposition. All the Mg-doped $In_xGa_{1-x}N$ layers show p-type conduction after thermal annealing. Room temperature (RT) carrier concentration increases exponentially with an In mole fraction increase. The highest hole concentration of bulk Mg-doped $In_xGa_{1-x}N$ is 1.65×10^{19} cm^{-3}. Also, the RT photoluminescence (PL) spectra of Mg-related emissions in $In_xGa_{1-x}N$ are displayed. However, the PL peak intensity becomes weak after the post-annealing process on Mg-doped $In_xGa_{1-x}N$. This degradation might be created by the surface dissociation during the post-annealing process.

Keywords: Mg-doped $In_xGa_{1-x}N$; Hall measurement; Carrier concentration; Mobility; In mole fraction; Photoluminescence

Group III nitrides are highly promising for applications in green to ultraviolet optoelectronic devices, due to their wide bandgap (1.9 eV of InN to 6.3 eV of AlN). Generally, for a conventional nitride optoelectronic and electrical device, the Mg-doped GaN is used for contact layer. However, self-compensation of Mg atoms in GaN films result in a low carrier concentration [1]. This creates an increased contact resistivity, which is not of suitable use as a contact layer. It is expected that p-type InGaN can be used as the contact layer in devices with low contact resistivity, because the bandgap is narrower than that of GaN [2]. Recently, Kumakura et al. demonstrated that p-type $In_xGa_{1-x}N$ [3] layer and high efficient hole concentration in Mg-doped InGaN/GaN superlattices [4]. In addition, they concluded that higher indium mole fractions created a higher hole concentration within Mg-doped $In_xGa_{1-x}N$ ($x < 0.2$). Moreover, much research has concentrated on the optical properties of undoped InGaN, and Zn-doped InGaN. However, there has been only a few reported cases of optical characters of Mg-doped InGaN [2].

In this study the electrical and optical characteristics of Mg-doped InGaN are investigated. The Mg-doped $In_xGa_{1-x}N$ layers have p-type conduction with a hole concentration that exceeds 10^{19} cm^{-3} (for $In_{0.23}Ga_{0.77}N$) by Hall measurements at room temperature (RT). As well, its photoluminescence (PL) spectra reveal Mg-related $In_xGa_{1-x}N$ emissions. However, the PL peak intensity becomes weakens after the post-annealing process on Mg-doped $In_xGa_{1-x}N$. This degradation might be caused by the thermal treatment during the post-annealing process.

Mg-doped GaN and $In_xGa_{1-x}N$ epitaxial layers were grown on a sapphire (0 0 0 1) substrate by low pressure metalorganic chemical vapor deposition (MOCVD). The growth was carried out at 100–300 torr with temperatures that ranged from 700°C to 900°C. Trimethylgallium (TMGa), Trimethylindium (TMIn), ammonia (NH_3) and bis-cyclopentadienylymagnesium (Cp_2Mg) were used as Ga, In, N, and Mg sources, respectively.

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Before growing the Mg-doped $In_xGa_{1-x}N$, a 30 nm thick low-temperature nucleation layer and an 2 μm undoped GaN were grown at 525°C and 1050°C, respectively. Their flow rates for each Mg-doped $In_xGa_{1-x}N$ films were equal. The growth temperature was controlled to vary the indium mole fraction in $In_xGa_{1-x}N$. The thickness of each Mg-doped $In_xGa_{1-x}N$ film varied from 1 μm (for GaN) to 0.13 μm (for $In_{0.23}Ga_{0.77}N$), which prevents the $In_xGa_{1-x}N$ layers from being cracked. After growth, the films were semi-insulated with resistivities of $>10^6$ Ωcm at 300 K.

The Mg-doped $In_xGa_{1-x}N$ films were then annealed in a nitrogen ambience at 750°C for 30 min. After annealing, each sample was characterized by double-crystal X-ray diffractometer, RT Hall effect and RT PL measurement. The indium mole fractions of $In_xGa_{1-x}N$ films were calculated by the difference, in X-ray rocking curve, of the peak position between $In_xGa_{1-x}N$ and GaN. These calculated values of the indium fraction in this study were ranged from 0 to 0.23. Hall measurements were taken by the van der Pauw method with Ni/Au ohmic contact [5]. PL was measured at RT via a 325 nm He–Cd laser.

Fig. 1 illustrates the RT carrier concentrations and mobility as functions of the $In_xGa_{1-x}N$ indium mole fraction. Notably, all $In_xGa_{1-x}N$ (x from 0 to 0.23) films showed p-type conduction after thermal annealing. For Mg-doped GaN films, RT carrier concentration was $3 \times 10^{17}cm^{-3}$ and mobility was 12 cm^2/V s. As we varied the growth temperature to increase the In mole fraction, the RT carrier concentrations increased exponentially with an In mole fraction increase. As the In mole fraction was increased, hole concentration saturation did not occur [3]. The hole concentration of Mg-doped $In_{0.23}Ga_{0.77}N$ was 1.65×10^{19} cm^{-3}. Notably, this concentration is the highest value for bulk Mg-doped $In_xGa_{1-x}N$ ever reported. Additionally, RT mobility decreased with an In mole fraction increase. This decrease further implied that RT mobility decreased with increased carrier concentration, and could be tentatively attributed to enhance scattering effect that results from the charged impurities.

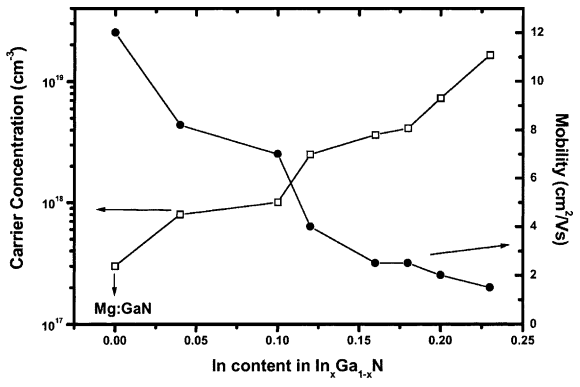

Fig. 1. RT carrier concentrations and mobility as functions of the $In_xGa_{1-x}N$ indium mole fraction.

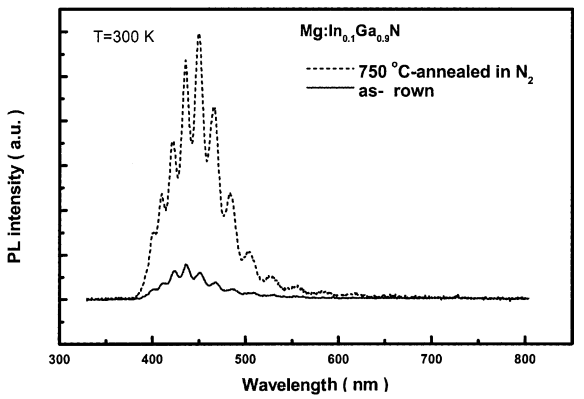

Fig. 2. RT PL spectra of the as-grown and activated Mg-doped $In_{0.1}Ga_{0.9}N$.

Fig. 2 demonstrates a typical RT PL spectrum of the as-grown and activated Mg-doped $In_{0.1}Ga_{0.9}N$. Both spectra showed oscillation caused by optical interference effect, thus indicating that the surface of this sample was specular. Alternately, the PL of the as-grown sample had a broader and weaker spectrum with peak wavelength at 435.7 nm and FWHM 49.5 nm. After the thermal annealing process, the peak intensity increased by one order of magnitude. This enhancement suggested that the significant activation of Mg acceptor in $In_{0.1}Ga_{0.9}N$ could be achieved by N_2-ambient thermal annealing. However the peak wavelength of activated $In_{0.1}Ga_{0.9}N$ shifts to 449.3 nm with FWHM 41.6 nm. This shift was owing to the identification deviation that was caused by optical interference.

Fig. 3 illustrates RT PL spectra of the as-grown and activated Mg-doped $In_{0.18}Ga_{0.82}N$. In Fig. 3 we can

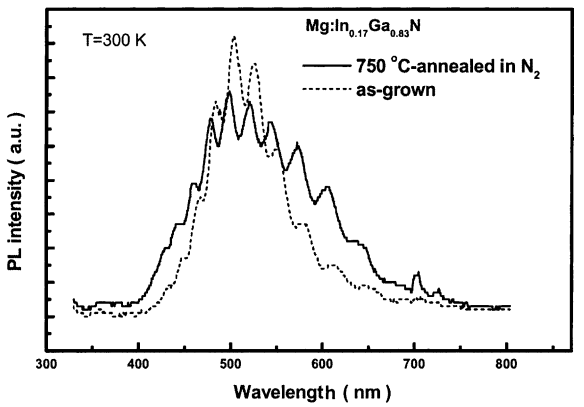

Fig. 3. RT PL spectra of the as-grown and activated Mg-doped $In_{0.}18Ga_{0.82}N$

clearly identify the optical interference effect as well as in Fig. 2. Further, the peak wavelength of the as-grown Mg-doped $In_xGa_{1-x}N$ was at 502.8 nm with FWHM 82.8 nm. However, the annealed sample had a weaker peak intensity and broader FWHM. The peak intensity of this annealed sample is 0.66 with peak wavelength at 498.3 nm and FWHM 158.6 nm. Interestingly, this trend differed from that of the PL characteristic of Mg-doped $In_{0.1}Ga_{0.9}N$. Therefore, we cannot identify whether Mg-doped $In_xGa_{1-x}N$ was activated after thermal annealing from PL spectra peak intensity.

Kumakura et al. reported that the activation energy for Mg-doped $In_xGa_{1-x}N$ decreased with an indium mole fraction increase [3]. Moreover, the acceptor concentration also increased with an indium mole fraction increase. This tendency was also noted herein. Fig. 1 illustrates that the carrier concentration increased exponentially with an indium mole fraction increase. Furthermore, the RT mobility of Mg-doped $In_xGa_{1-x}N$ decreased with an indium mole fraction increase. This implies that with an increased carrier concentration, mobility decreased, which also corresponds with the "ionized impurity scattering" [6] mechanism. Restated, as the impurity concentration increases, the number of impurity scattering centers increase, therefore mobility is reduced.

The In mole fraction of the $In_xGa_{1-x}N$ films was calculated from the difference of X-ray diffraction peak position between $In_xGa_{1-x}N$ and GaN. From the In mole fraction, the bandgap energy of $In_xGa_{1-x}N$ can be determined by following equation [7]

$$E_g(x) = 3.50(1-x) + 1.97x - 2.6x(1-x) \quad (\text{eV})$$

Thus, the bandgap energy of $In_{0.1}Ga_{0.9}N$ and $In_{0.18}Ga_{0.82}N$ are 3.11 and 2.84 eV, respectively. However, the band-edge emission of InGaN in PL spectra was not observed. Instead, longer wavelength peaks were observed, which was thought to be an Mg-related emission. A comparison of Figs. 2 and 3 revealed that, with an In mole fraction increase, the Mg-related emission shifted towards the longer wavelength [2]. Moreover, Nakamura et al. indicated that the Zn-related emission of $In_xGa_{1-x}N$ was between 0.4 and 0.5 eV lower than the band-edge emission energy [8]. The difference between the estimated band-edge and Mg-related emission herein was 350 meV for $In_{0.1}Ga_{0.9}N$ annealed sample and 374 meV for $In_{0.18}Ga_{0.82}N$ annealed sample. That is, the Mg-related emission of $In_xGa_{1-x}N$ was approximately 350 and 375 meV lower than the band-edge emission energy. Therefore, one can tentatively estimate the Mg-related emission in Mg-doped $In_xGa_{1-x}N$ by the above mentioned relationship.

A comparison of the PL peak intensity of both the as-grown and annealed sample in Fig. 2 confirms that the peak intensity increased by one order of magnitude after annealing. This indicates that, following thermal annealing, Mg atoms in InGaN were activated. Similar results had also been addressed to Mg-doped GaN [8]. However, the peak intensity of the annealed sample was weaker than the as-grown one, high In-content samples especially. This reduced intensity might due to the excessive surface dissociation that occurred during the annealing process. As stated previously, the growth temperature of Mg-doped $In_xGa_{1-x}N$ ranged from 700°C to 800°C. Notably, the $In_{0.1}Ga_{0.9}N$ growth temperature (780°C) was higher than the annealing temperature (750°C). Thus, this sample did not degrade during thermal annealing. In contrast, the $In_xGa_{1-x}N$ growth temperature was 740°C, which was lower than the annealing temperature. Therefore, optical degradation might occur during the annealing process. Further investigation is required to determine the optimum annealing condition.

In summary, the electrical and optical properties of Mg-doped $In_xGa_{1-x}N$ were investigated herein. All the Mg-doped $In_xGa_{1-x}N$ layers had p-type conduction. As well, with an In mole fraction increase, the RT carrier concentration increased exponentially. The hole concentration of Mg-doped $In_{0.23}Ga_{0.77}N$ was 1.65×10^{19} cm^{-3}, which is the highest value for bulk Mg-doped $In_xGa_{1-x}N$ reported previously. Also, the RT PL spectra of the Mg-related emission in $In_xGa_{1-x}N$ were displayed. However, the PL peak intensity weakened after the post-annealing process on Mg-doped $In_xGa_{1-x}N$ with high In composition. This degradation might have been caused by the excessive surface dissociation during the post-annealing process.

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