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Heteroepitaxial growth of InN on GaN intermediate layer by PA-MOMBE

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ABSTRACT

In this paper, high-quality wurtzite indium nitride was epi-grown on sapphire substrates by plasma-assisted metal-organic molecule beam epitaxy system (PA-MOMBE). Structural and electrical properties of the InN films were significantly improved by employing a GaN buffer layer. In addition, high-resolution X-ray diffraction, field emission scanning electron microscopy, transmission electron microscopy (TEM), Hall Effect, Raman and photoluminescence spectroscopy were carried out to characterize the effect of the growth temperature on structural and optoelectronic properties. It was found that highly *c*-axis oriented InN epilayer can be obtained by optimizing growth conditions. TEM images reveal that the epitaxially grown InN/GaN interface is sharp, and the spacing of the InN(0 0 0 2) lattice plane is about 0.57 nm. Raman spectra also show a sharp peak at 491 cm^{-1} attributed to the E_2(high) mode of wurtzite InN. These results indicate that the improvement of InN material quality can be achieved using heteroepitaxy on GaN/sapphire templates.

1. Introduction

Due to a direct bandgap of approximately 0.65 eV and a high electron saturation velocity, indium nitride (InN) has attracted much attention as a potential material for device applications such as solar cells, optoelectronics and high-frequency devices [1–5]. Despite these potential applications, InN is the least understood material among the III-nitride compounds because of a low InN decomposition temperature, 500–600 °C [6]. Under such a low growth temperature, the surface migration of the precursors may be insufficient for the InN growth with conventional growth techniques [7,8], such as metalorganic chemical vapor deposition (MOCVD) and molecular beam epitaxy (MBE). Besides, lowering the growth temperature often tends to increase defect and dislocation densities, and results in the disorder-related high carrier concentrations. Ammonia (NH_3) is commonly used as a nitrogen source for the growth of nitrides in metalorganic vapor phase epitaxy and the pyrolysis efficiency of NH_3 is very low for temperatures lower than 650 °C. On the other hand, since bulk InN substrates are not available, sapphire (a-Al_2O_3) is the most used substrate material for the heteroepitaxy InN growth. However, epitaxial growth of InN directly on *c*-plane sapphire was not successful because of the large lattice mismatch (~25%). Thus, introducing suitable buffer was thought to be a suitable alternative for high-quality InN growth [9–13].

Here, we present the heteroepitaxal growth of InN on GaN intermediate layer by plasma-assisted metalorganic molecular beam epitaxy (PA-MOMBE), which can effectively provide more active nitrogen species to overcome the low dissociation rate occurred in the MOCVD system at low growth temperature. By carefully using plasma-assisted growth techniques, we were able to grow high-quality InN epifilms at a moderate temperature. Photoluminescence (PL) measurement was conducted to characterize the thermal quenching mechanism of the heteroepitaxial InN/GaN film. In addition, structural and electrical properties of InN thin films grown on GaN intermediate layers were investigated.

2. Experimental procedure

Undoped InN epilayers were grown in a home-made PA-MOMBE on GaN-epilayer/sapphire(0 0 0 1) templates. Trimethylindium (TMI) was used as the In precursor source, and the atomic nitrogen generated by radio-frequency (RF) plasma was the group-V source. The undoped 5 μm thick GaN templates were grown by a conventional two step growth method and exhibited a high crystal quality: Hall concentration of $\sim 1\times 10^{17}$ cm^{-3}, Hall mobility of

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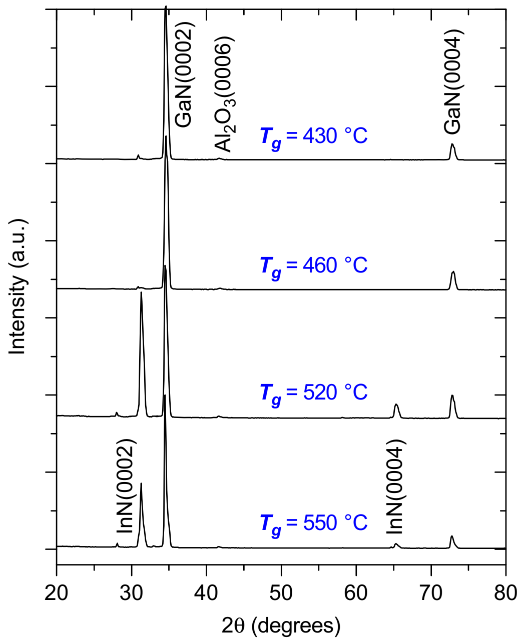

Fig. 1. Normalized $\omega-2\theta$ XRD patterns of InN film deposited on GaN/sapphire templates with different growth temperatures (T_g).

$\sim$300–400 cm^2/V s, and the full-width at half-maximum (FWHM) for GaN(1 0 2) X-ray diffraction (XRD) of $\sim$380 arcsec.

The growth of InN was performed in a PA-MOMBE chamber with a background pressure of 10^{-9} Torr. N_2 gas (6 N purity) was introduced into the growth chamber through a RF-plasma radical source operated at 350 W, while the pressure of the chamber was kept at 1.5×10^{-5} Torr during the InN growth. By varying the growth temperature, the optimal growth temperature (T_g) for InN epilayers deposited on GaN templates was found to be around 520 °C and InN films were observed to decompose or evaporate as T_g was raised to above 520 °C. During the deposition, the substrate temperature was monitored by a thermocouple and regulated by the IR pyrometer with a PID programmable heater.

The X-ray diffraction (BeDe D1) measurements have been carried out in a $\omega-2\theta$ coupled geometry using Cu-$K\alpha$ radiation to investigate structural properties. Field-emission scanning electron microscope (Hitachi S-4300 FE-SEM) was used to examine the surface and cross-section morphologies of the InN films. The microstructure of the InN films was investigated in detail by TEM in cross-section configuration (TEM, Philips Tecnai 20). Electrical properties of the films were investigated by room temperature Hall effect measurement. Photoluminescence (PL) measurements were performed at 14 K in a closed-circuit helium refrigerator. The luminescence was dispersed by a 0.55 m monochromator and collected by an InGaAs photodetector interfaced with a lock-in

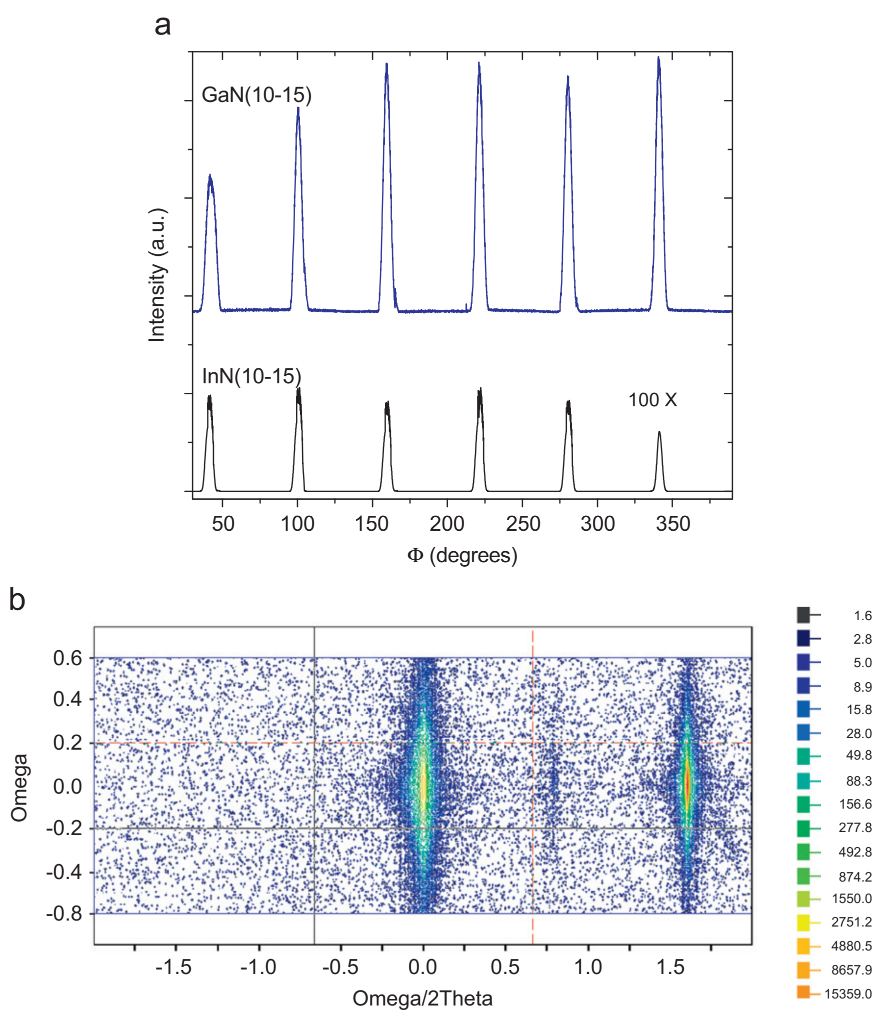

Fig. 2. (a) XRD ϕ scans of the InN(1 0 −1 5) and GaN(1 0 −1 5) peaks of InN film hetero-epitaxially grown at 550 °C, and (b) diffraction space mapping of ω versus $\omega/2\theta$ around InN(0 0 0 2) and GaN(0 0 0 2) peaks.

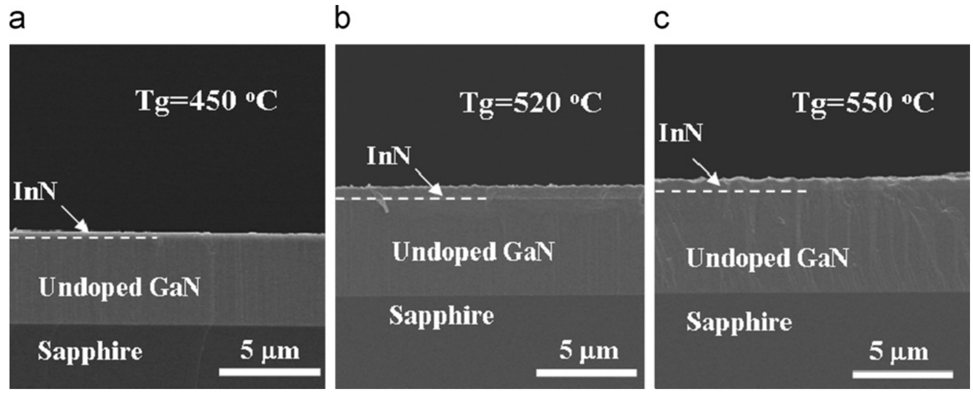

Fig. 3. Cross-sectional SEM images of InN films grown at (a) 450 °C, (b) 520 °C and (c) 550 °C.

amplifier with a diode-pumped solid state laser emitting at 532 nm was used for excitation.

3. Results and discussions

Fig. 1 shows normalized $\omega-2\theta$ scan (conventional $\theta-2\theta$ scan, radial scan) XRD spectra of InN heteroepitaxial layers with different growth temperatures. It is obvious that the XRD diffraction intensities of InN films increased with a gradual increase in growth temperatures. On the other hand, InN films were observed to decompose as T_g was raised to above 550 °C. Several diffraction peaks: InN(0 0 0 2), InN(0 0 0 4), GaN(0 0 0 2) and Al_2O_3(0 0 0 6) were indexed, and this is indicative of [0 0 0 1] oriented hexagonal InN, which was epitaxially grown on GaN/sapphire template. The *c*-axis lattice constant of the obtained InN thin films was calculated to be 5.69 Å utilizing the observed InN(0 0 0 2) diffraction peak, which is slightly larger than the bulk value ($\sim$5.67 Å). The phenomenon indicates the InN films grown on GaN/sapphire templates are tensile strained in the *c*-axis direction.

In order to determine the epitaxial relationship between InN and underlying GaN epilayers, the ϕ scan of the InN(1 0 −1 5) and reciprocal space mapping (RSM) covering the GaN(0 0 0 2) and the InN(0 0 0 2) diffraction peaks are adopted. The ϕ scans of the InN(1 0 −1 5) and GaN(1 0 −1 5) peaks are presented in Fig. 2(a). The hexagonal structure of InN produces six equal-spaced peaks. Absence of any other random peaks suggests that the InN film grains predominantly grow in the direction of [0 0 0 1], and the 0° shift between the InN and GaN is explained by coherent in-plane growth. The reciprocal space mapping (RSM) of the grown structure was obtained as shown in Fig. 2(b). It is observed that the InN layer grows coherently on the GaN since there is minimal tilting between the plane of InN and GaN. The smaller FWHM in the $\omega-2\theta$ direction and the relatively large FWHM in the ω direction imply that the structural properties are most likely influenced by dislocations rather than compositional variation in the epilayer. A noticeable feature of the InN(0 0 0 2) peak is that it is broadened along the ω direction, indicating the presence of high density dislocations in this film.

The cross-sectional SEM images of the InN films are shown in Fig. 3. The interfaces between InN, GaN and sapphire are clearly visible. Meanwhile, the InN exhibits a pronounced characteristic of two-dimensional growth mode at lower T_g. While increasing T_g, the vertical growth might become favored over lateral growth and thus the surface morphology became rougher as seen in Fig. 3(c). Besides, the InN growth rate varied from 0.72 to 1.78 µm/h with an increase in growth temperature.

To further confirm the structural properties of the heteroepitaxial growth of InN on GaN intermediate layer, TEM measurement

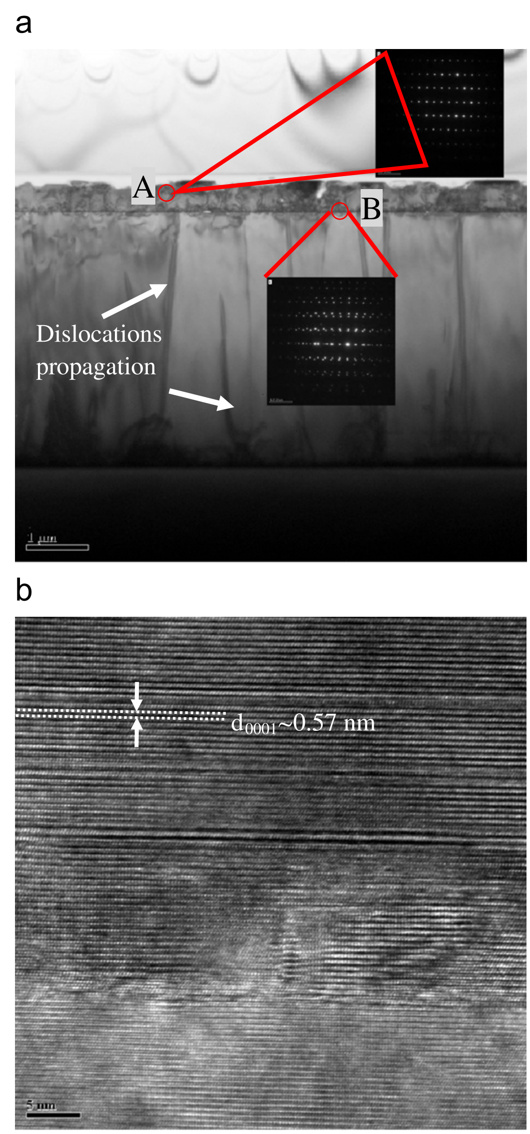

Fig. 4. (a) Cross-sectional HRTEM images of InN film epi-grown at 550 °C. Insets are the SAED patterns of InN layer and InN/GaN interface, labeled as A and B. (b) Lattice images of InN epi-film and the lattice constant is about 0.57 nm.

was employed. Fig. 4(a) shows the cross-sectional TEM image of InN epilayers grown at 550 °C. It is noteworthy that most threading dislocations (TDs) in the GaN are blocked out in the InN/GaN interface. Thus, most of the line defects in InN epilayers likely originated from the film deposition process or the InN/GaN interface. Shown in the insets are the selected area electron diffraction (SAED) patterns of InN and InN/GaN interface labeled as A and B. The SAED patterns indicated that the orientation relationship between the InN films and GaN intermediate epilayer is InN(0 0 0 1) parallel to GaN(0 0 0 1), consistent with the HRXRD results. Furthermore, no extra diffraction spots were observed in area B. This observation implies that no interlayer exists between InN and GaN. Fig. 4(b) is a HRTEM image of the InN/GaN interface. Stacking faults and/or dislocations were observed in the InN films. The lattice constant of InN epilayer grown at 550 °C is about 0.57 nm.

Further enhancement in the electrical properties of InN on GaN/sapphire template was obtained by increasing the growth temperature. Fig. 5 shows the dependence of the carrier concentration (n) and mobility (μ) of InN on the T_g. It is seen that n decreases from 3×10^{20} to 2×10^{19} cm^{-3} and μ increases from ~30 to ~400 cm^2/V s as the growth temperature increases from 450 to ~550 °C. The high carrier concentration might result from nitrogen vacancies and defects due to lattice mismatch. Improved crystal quality by introducing GaN intermediate epilayer is the plausible explanation of the decreased carrier concentration. It is evident that high growth temperature decreased the carrier concentration of the InN epilayer. The origin of the background carrier concentration has been attributed to a native donor of nitrogen vacancy (V_N), an extrinsic donor of oxygen and other structural defects. As seen earlier, it has been found that the heteroepitaxial InN film can be structurally improved through reduction of lattice mismatch to the underlying layer by inserting GaN intermediate layer at the moderate growth temperature. Therefore, 10 times enhancement of carrier mobility is reasonable by modulating the growth temperature.

To evaluate optical quality of the InN epifilms, temperature-dependent PL was measured and presented in Fig. 6. The asymmetric PL spectra may result from surface electron accumulation and absorbed moisture in InN films. As the temperature increases the PL intensities gradually decreased. Previous literatures have reported that the carriers could receive activation energy to thermalize from potential minima to nonradiative or delocalized centers as the temperature was increased. To get insight into the thermal quenching process associated with the localized states, the PL intensity as a function of temperature was studied, and the experimental temperature-dependent PL data were fitted by Arrhenius equation to investigate the carrier behavior during

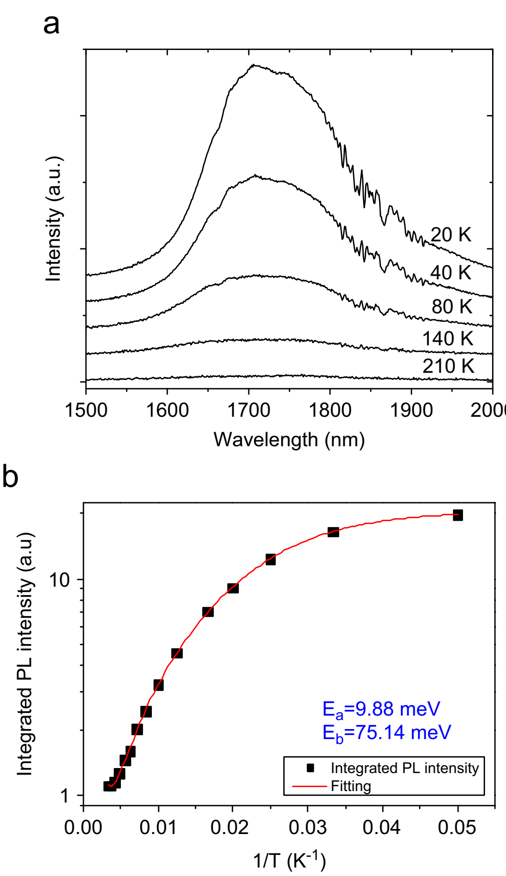

Fig. 6. (a) Temperature-dependent photoluminescence spectra of InN epi-film, and (b) PL integrated intensity versus 1/T; dotted line: Arrehnius plot.

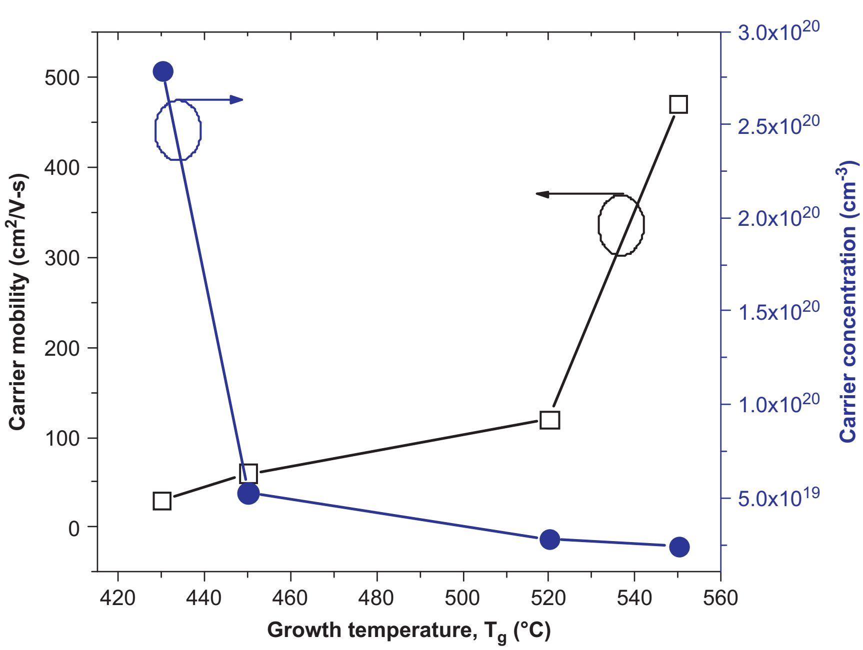

Fig. 5. Carrier mobility and concentration of InN grown on GaN/sapphire templates with growth temperature.

the thermal processes [14]

$$I(T) = \frac{I_0}{1+A\exp(-E_a/K_BT)+B\exp(-E_b/K_BT)},$$

where I_0 is the PL intensity at low temperature, K_B the Boltzmann's constant, the coefficients $A1$ and $A2$ measure the strengths of both quenching mechanisms, and $E1$ and $E2$ are the thermal activation energies in the high- and low-temperature regions, respectively. The solid lines in Fig. 6(b) show the fitted results, which agree closely with the experimental data. We suggest that E_a is caused by the nonradiative channel through defects or dislocations in InN, and E_b represents the localization energy of the localized carriers. However, further understanding concerning the exact mechanisms is needed and underway.

4. Conclusions

In conclusion, we have successfully grown high-quality InN epilayer on GaN/sapphire template by PA-MOMBE. By adjusting the growth temperature, it is evident that high growth temperature decreased the carrier concentration of the InN films, and thus the carrier mobility can be effectively increased by one order of magnitude. The XRD and SAED patterns indicated that the orientation relationship between the InN films and GaN intermediate epilayer is InN(0 0 0 1) parallel to GaN(0 0 0 1). Moreover, HRTEM images reveal that the epitaxially grown InN/GaN interface is sharp. Temperature-dependent PL analysis implies existence of two nonradiative recombination channels. These results indicate that the improvement of InN material quality can be achieved using heteroepitaxy on GaN/sapphire templates. Further research is underway to clarify the quench mechanism in InN films heteroepitaxy on GaN intermediate layer.

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