

# Structural and Morphological Characteristics of $\text{In}_x\text{Ga}_{1-x}\text{N}$ Films Grown on Si (111) by Reactive Magnetron Sputtering

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**Abstract.** Under various power ratios and temperatures,  $\text{In}_x\text{Ga}_{1-x}\text{N}$  films with different indium composition  $x$  were grown on Si(111) substrates by reactive magnetron sputtering, and then annealed at ammonia atmosphere around 700°C for 2 hours. The indium composition  $x$  can be adjusted by the growth temperature over the range of 600–800°C. There is no InN phase and In droplet formation in the  $\text{In}_x\text{Ga}_{1-x}\text{N}$  films due to the low-temperature advantages of reactive magnetron sputtering. The rich In composition in  $\text{In}_x\text{Ga}_{1-x}\text{N}$  films is caused by the higher sputtering yield of  $\text{In}_2\text{O}_3$  target than  $\text{Ga}_2\text{O}_3$  target. Raman scattering analysis revealed that the  $\text{In}_x\text{Ga}_{1-x}\text{N}$  films obtained at different temperatures were wurtzite structure, and the compositional inhomogeneity is caused by the relaxation of momentum conservation and increase of lattice disorder.

## 1 Introduction

Recently, the III-nitride semiconductor materials has attracted significant attention on account of its outstanding performance applied in ultraviolet light-emitting diodes and laser diodes, etc.  $\text{In}_x\text{Ga}_{1-x}\text{N}$  has been widely studied owing to its efficient photovoltaic applications. While adjusting the In composition ratio, the band gap energy of  $\text{In}_x\text{Ga}_{1-x}\text{N}$  is tunable from 0.7eV to 3.4eV, which covers full solar spectrum[1]. However, it is a technical bottleneck to grow  $\text{In}_x\text{Ga}_{1-x}\text{N}$  thin film with well crystallization quality due to the obvious

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lattice mismatch between InN and GaN[2]. Traditional techniques to prepare  $\text{In}_x\text{Ga}_{1-x}\text{N}$  film including MOCVD, MBE and MOVPE, which need very high cost. In our work,  $\text{In}_x\text{Ga}_{1-x}\text{N}$  films are grown by reactive sputtering technique.

## 2 Experimental

In the present work, the films were prepared by a reactive radio-frequency magnetron sputtering system (MSP-3200C2).  $\text{Ga}_2\text{O}_3$  was chosen as gallium source, and  $\text{In}_2\text{O}_3$  as indium source. After the pretreatment of substrates, the first set of  $\text{In}_x\text{Ga}_{1-x}\text{N}$  was performed at growth temperature of  $600^\circ\text{C}$ , and the sputtering power ratios of  $\text{In}_2\text{O}_3$  and  $\text{Ga}_2\text{O}_3$  were 1:1(100W:100W) and 1:2(75W:150W) respectively. The second set of the experiment grew at  $600\sim 800^\circ\text{C}$  at the same sputtering power ratio of 1:1. The sputtering lasted for 30 min at the concentration of 50%  $\text{N}_2$  with the pressure of 0.55 Pa. Then, the obtained films of two groups in pipe furnace (TCW-32B) have been processed to get the  $\text{In}_x\text{Ga}_{1-x}\text{N}$  films with stable ammonia flow (500ml/min) at  $700^\circ\text{C}$  for 2 hours. The crystal structure of the samples were characterized by X-ray diffraction (XRD, 6100, SHIMADZU). The morphology and composition of the samples was examined by Scanning Electron Microscope (SEM, Zeiss SIGMA) and Energy Diffraction Spectrum (EDS). Raman microspectrometer is used a He-Ne laser with the excitation wavelength of 633nm.

## 3 Results and discussion

Fig.1 shows the XRD patterns of films deposited on Si(111) substrates under various sputtering power ratios 1:1 (100W:100W) and 1:2(75W:150W) of  $\text{In}_2\text{O}_3$  and  $\text{Ga}_2\text{O}_3$ . (002), (100) and (101) peaks of  $\text{In}_x\text{Ga}_{1-x}\text{N}$  films are clearly seen in the range of InN and GaN peaks with wurtzite structure. The results indicate that the deposited films are ternary alloy. There is no InN phase and In droplet formation at the  $\text{In}_x\text{Ga}_{1-x}\text{N}$  films, which manifested that the films has single crystal phase.

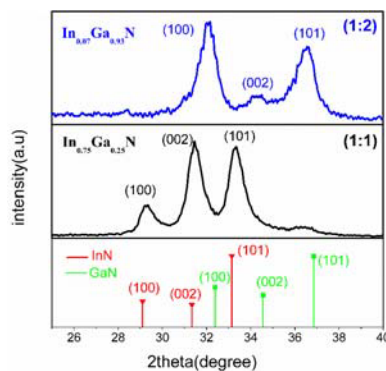


Fig. 1 XRD patterns of the  $\text{In}_x\text{Ga}_{1-x}\text{N}$  thin films at different power ratios

Fig.2 shows the SEM and EDS analysis of  $\text{In}_x\text{Ga}_{1-x}\text{N}$  films at different power ratios. In Fig.2(a), there are a few crystal grains at the surface, less than in Fig.2(b). Since the energy supplied by RF-power(1:2) was less than the sputter power ratio of 1:1, fewer atoms were sputtered from targets and the reaction of atoms at substrate surface decreased greatly. The In/(Ga+In) ratios were 0.07 and 0.75 under the sputtering power ratios of 1:2 and 1:1 respectively, which demonstrates that the sputtering yield of In is much higher than Ga and is the same as Cheng-Che Li et al[3]. In words, it denotes that the RF-power ratio of 1:1 is suitable for obtaining In-rich alloy films.

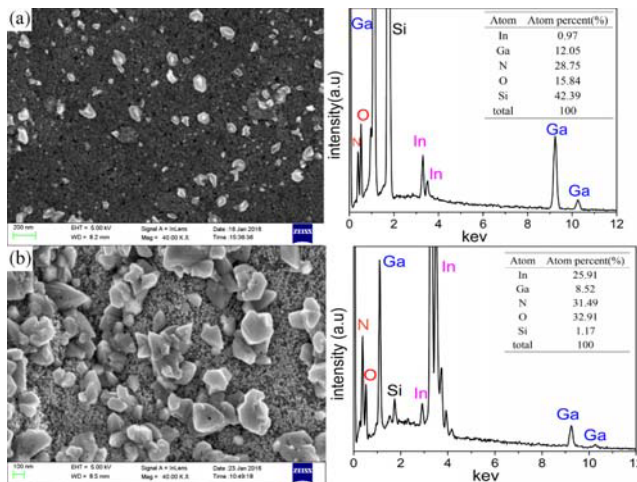


Fig. 2 SEM and EDS composition analysis of  $\text{In}_x\text{Ga}_{1-x}\text{N}$  films of power ratio of 1:2(a) and 1:1(b)

The corresponding XRD spectra of the second set of films is given in Fig.3. It is clearly observed that the  $2\theta$  angular position shifts towards higher values while the temperature increases, meanwhile In composition  $x$  decreases, which is the same as [4] and [5]. It was caused by the process of In adsorption and desorption in deposition, the suppression of In desorption reserves more In atoms in films at lower temperature. When the temperature increases, In-N bond begins to split and In atoms will divorce from the film surface because the evaporated temperature of In is around  $700^\circ\text{C}$  or more.

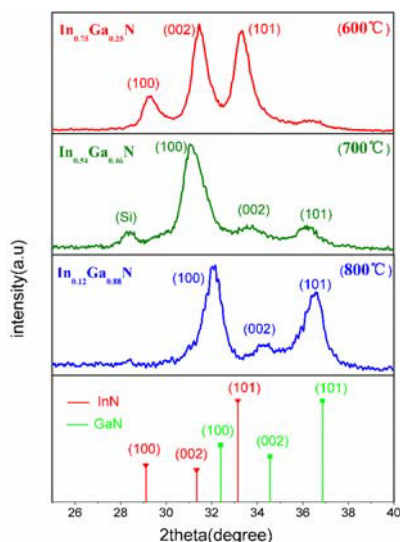


Fig. 3 XRD patterns of the In<sub>x</sub>Ga<sub>1-x</sub>N thin films at different growth temperature

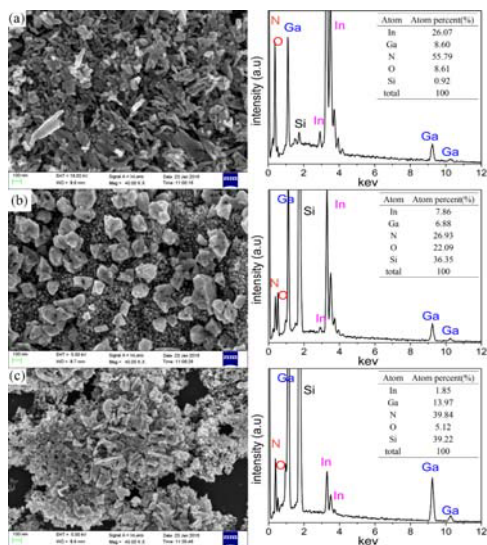


Fig. 4 SEM images and EDS composition analysis of In<sub>x</sub>Ga<sub>1-x</sub>N films at temperatures of (a)600°C, (b)700°C and (c)800°C

The SEM morphology of In<sub>x</sub>Ga<sub>1-x</sub>N samples with different temperature were shown in Fig. 4. It presented linear columnar and block large particles and schistose at 600°C, 700°C and 800°C respectively, and the In/(Ga+In) ratios were 0.75,0.54,0.12 at corresponding temperature, which may be caused by defect of the metal In evaporated under different temperature.. From the EDS composition result, no matter how to set the growth parameter, O impurity always existed in the film, which may come from target. The thickness varies with temperatures and the rate of high temperature ammoniating is different, hence the content of oxygen is different in films. In conclusion, the temperature between 600°C and 700°C is suitable for obtaining In-rich alloy films.

Fig. 5 shows Raman spectra with different In composition at different temperatures. The spectra was recorded in the backscattering geometry configuration with a He-Ne laser at 633nm. InN and GaN are both  $C_{6v}^4$  point group symmetry structure (belong to wurtzite structure) [6], which is represented by  $A_1(\text{LO})$  and  $E_2(\text{high})$  modes. The silicon substrate peak at  $520\text{cm}^{-1}$  is so strong that makes the  $E_2(\text{high})$  weakened. The  $A_1(\text{LO})$  mode of three samples in different temperatures are observed in the region  $584\text{--}612\text{cm}^{-1}$  which refers that the  $A_1(\text{LO})$  mode of InN located in  $570\text{cm}^{-1}$  and  $733.07\text{cm}^{-1}$  for GaN[7]. The peak positions move towards lower wavenumbers while the In compositions  $x$  increases, the main reason for this shift is structural defects in films. The  $A_1(\text{LO})$  mode of  $\text{In}_x\text{Ga}_{1-x}\text{N}$  films with different In compositions manifested that the alloy films are wurtzite structure[8]. The rough curve of the main board band in the region  $584\text{--}612\text{cm}^{-1}$  can attribute to the relaxation in momentum conservation and increasing lattice disorder due to the compositional inhomogeneity[9].

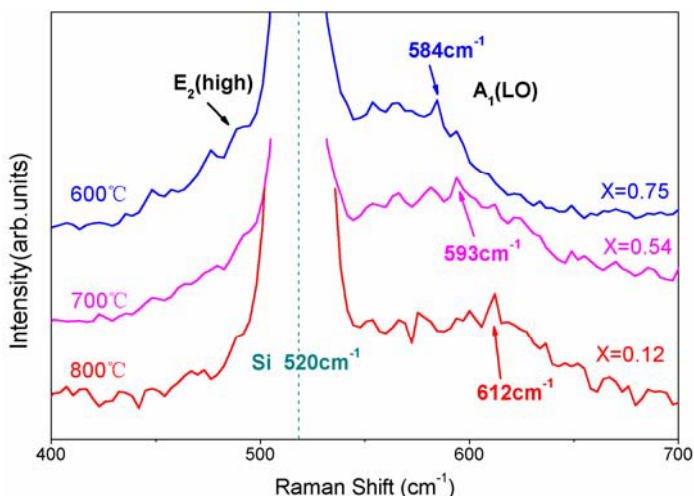


Fig. 5 Raman spectra of  $\text{In}_x\text{Ga}_{1-x}\text{N}$  films at different indium composition  $x$

## 4 Conclusion

In this work, we have grown  $\text{In}_x\text{Ga}_{1-x}\text{N}$  of different indium composition  $x$  at variation of power ratios and temperatures by reactive magnetron sputtering. It was demonstrated that the indium composition  $x$  was not linear with power ratios, which was caused by the high sputtering yield of the  $\text{In}_2\text{O}_3$  target. The monotonic decrease in  $x$  is caused by the more In evaporation with increasing temperatures. Raman scattering analysis revealed that the reasons for the shift of  $A_1(\text{LO})$  phonon in the region  $584\text{--}612\text{cm}^{-1}$  are In composition variation. The results of the experiment also implied that pure  $\text{In}_x\text{Ga}_{1-x}\text{N}$  alloy film in reactive magnetron sputtering may apply in development optoelectronic devices.

## 5 Acknowledgment

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