

Marked improvements in electrical and optical properties for MOVPE InN annealed at a low temperature (300°C) in O₂ atmosphere

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We have found that the material quality of MOVPE InN can be markedly improved after the annealing in the air at around 300°C. By the annealing in the air, carrier concentration is reduced by about one order of magnitude. In accordance with the carrier reduction, PL intensity is increased and PL peak energy is shifted to the lower energy side by about 0.06 eV for the film annealed for 3h. The reduction of carrier concentration is also conformed by the shift of LO phonon-plasmon coupled mode in the Raman spectrum. The FWHM of the E₂(high) mode is decreased, indicating that the crystalline quality is slightly improved by the annealing. Since the FWHM of X-ray rocking curve is not changed after the annealing, the improvement by the annealing is concluded not to be in macroscopic scale but microscopic scale. No improvements are found for the samples annealed in the N₂ flow. No data that show the chemical oxidation of InN are also found.

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1 Introduction

Although indium nitride (InN) is still a less studied material than other III-nitride semiconductors, it is expected to have the smallest effective mass, 0.07m₀[1]-0.14m₀ [2], and the highest electron drift velocity, 4.2 x 10⁷ cm/s [3], in III-nitrides. Therefore, InN is expected to be a channel material for high-speed and high-frequency electron devices. In order to accomplish these applications, considerable improvements in electrical/optical and crystallographic improvements will be required for InN films. Presently, a carrier concentration in the order of 10¹⁷ cm⁻³ has been realized for MBE InN, while that for MOVPE InN has been still in the range 10¹⁹-10¹⁸ cm⁻³. Therefore, it is highly desirable to clarify the causes for the higher residual carrier concentration in MOVPE InN.

We have found for the first time that electrical and optical properties for MOVPE InN are markedly improved when the samples are annealed in the oxygen atmosphere at a low temperature around 300 °C. This paper reports the electrical and optical properties for MOVPE InN annealed at 300°C in the air, in comparison with those for samples annealed in the N₂ flow.

2 Experimental

Using a MOVPE system with a horizontal reactor, an InN film with a thickness about 0.5 μm is grown at 600 °C on (0001) sapphire substrates with a GaN buffer. After the growth, InN samples are annealed at 300°C for 3-10 h in the air or in the N₂ flow (6 slm). For samples before and after the annealing, carrier concentration and photoluminescence (PL) spectrum are measured at room temperature. Carrier concentration is measured with the Hall effect with the van der Pauw method. Using a He-Cd laser (442 nm) as an excitation source and a liquid nitrogen-cooled InGaAs pin photodiode as a detector, PL spectrum is measured from the front surface or the back surface of an InN film through the sapphire substrate. Structural characterization is also made with X-ray diffraction. Raman spectra are measured at RT using a Raman microscope in the back-scattering geometry from the grown surface, with a 458 nm Ar⁺ laser.

3 Results and discussion

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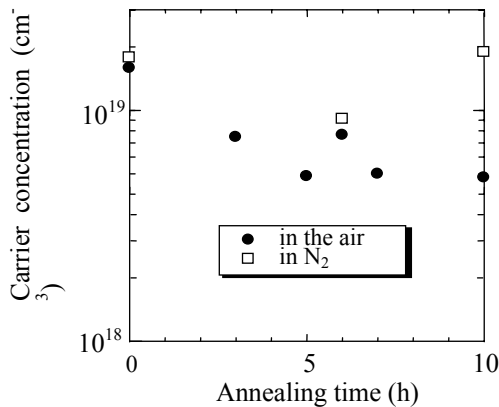


Fig.1. Annealing time dependence of carrier concentration for InN films annealed at 300°C in the air or N₂ atmosphere.

Figure 1 shows the carrier concentration for InN as a function of post-growth annealing time. By annealing the samples at 300°C in the air for 5 h, the carrier concentration is reduced from 2×10^{19} to 5×10^{18} cm⁻³. The carrier concentration of 5×10^{18} cm⁻³ corresponds to the lowest one obtainable for MOVPE InN. No significant change is brought by the annealing in the N₂ atmosphere. From these facts, we can conclude that the annealing in the oxygen ambient have a passivation effect of donors in InN. No marked change in electron mobility is found after the annealing in the air. This means that the mobility is not governed by the ionized donors in MOVPE InN [4]. Figure 2 shows the PL peak energy measured at room temperature as a function of post growth annealing time. PL spectra are measured with

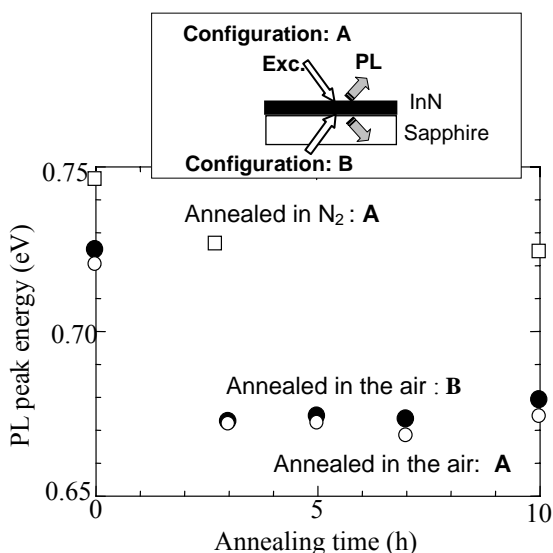


Fig. 2. Annealing time dependence of PL peak energy for InN films annealed at 300°C in the air or N₂ atmosphere. PL spectrum is measured with the two different configurations, i.e., from the front (configuration A) or the interface side (configuration B) of the films.

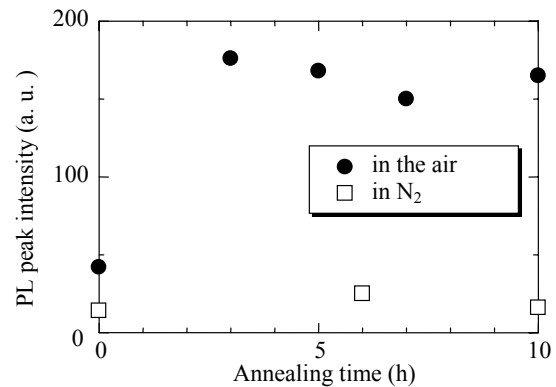


Fig. 3. Annealing time dependence of PL intensity for InN films annealed at 300°C in the air or N₂ atmosphere.

the two different configurations, i.e., from the front (configuration A) or the back surfaces (configuration B) of samples [5]. When the annealing is made in the air, PL peak energy is decreased from 0.73 eV for the as-grown sample to 0.67 eV for the film annealed for 3h. The PL peak energy of 0.67 eV corresponds to the lowest one obtainable for MOVPE InN. The reduction of carrier concentration and the decrease of PL peak energy can be explained by the Burstein-Moss effect. One can see that there is no difference between the PL peak energies measured from the back and front surfaces. This means that such an improvement occurs in the whole part of the film. The change in PL peak energy for the samples annealed in the N₂ is very small as seen in Fig. 2. Figure 3 shows the change in PL intensity after the annealing. As seen in Fig. 3, the PL intensity increased by the factor of about 3 when the sample is annealed for 3 hours. The results shown in Figs. 2 and 3 clearly show the optical improvement for the InN films annealed in the air. Figure 4 shows the Raman spectra for as-grown and annealed InN films. One can see that with increasing annealing time, LO phonon-plasmon coupled mode is shifted from 440 cm⁻¹ to 430 cm⁻¹. This shows the

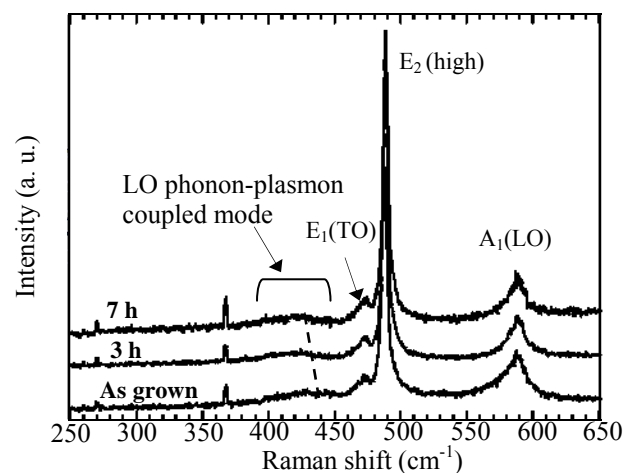
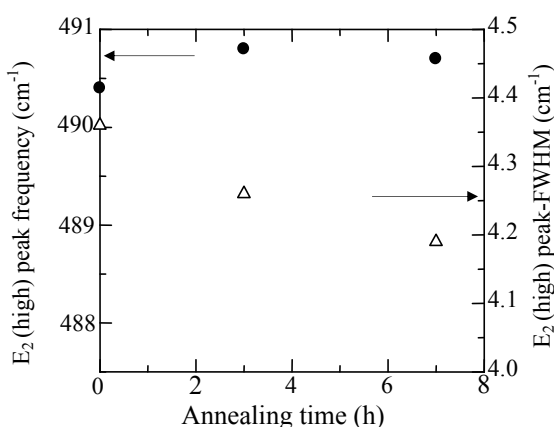


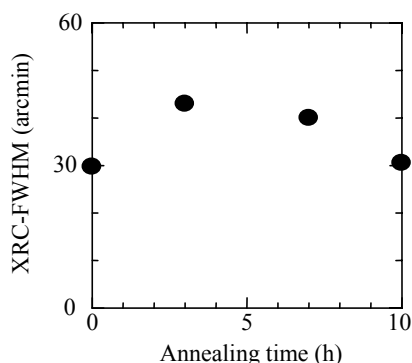
Fig. 4. Raman spectra for as-grown and annealed InN films.

1 decrease of carrier concentration, supporting the Hall and
 2 PL improvements shown in Figs. 1-3. It should be noted
 3 that for all the samples, no In_2O_3 -related peaks are found.
 4 This means that no chemical oxidation occurs in this case.
 5 Figure 5 shows the annealing time dependence of the
 6 Raman shift and FWHM of the E_2 stress in InN is not
 7 changed by the annealing. As seen in Fig. 5, FWHM of the
 8 E_2 (high) mode is slightly decreased. (high) mode. No shift
 9 of the E_2 (high) is observed for the annealed samples.
 10 Therefore, one can see that the residual
 11 Therefore, the crystalline quality of InN film is slightly
 12 improved by the annealing in the air. Figure 6 show the
 13 annealing time dependence of X-ray rocking curve (XRC)
 14 for InN films annealed at 300°C in the air. It is clear that
 15 the XRC-FWHM is not changed by the annealing in the air.
 16 Therefore, it is concluded that the improvement of crystal-
 17 line quality for InN by the annealing is not in macroscopic
 18 scale but microscopic scale.

19 The cause for the improvements of electrical and optical
 20 properties found in this study is not clear at present. One of



37 Fig. 5. Annealing time dependence of E_2 (high) peak
 38 frequency and FWHM.



54 Fig.6. Annealing time dependence of FWHM of
 55 (0002) X-ray rocking curve for InN films annealed
 56 at 300°C in the air.
 57

the probable reasons may be the oxygen passivation of do-
 nors such as hydrogen incorporated during the growth.
 Further investigations will be needed to clarify the mecha-
 nism of the improvements.

4 Conclusion

We have found that the material quality of MOVPE
 InN can be markedly improved after the annealing in the
 air at around 300°C. By the annealing in the air, carrier
 concentration is reduced from 2×10^{19} to $5 \times 10^{18} \text{ cm}^{-3}$. In
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 Since the FWHM of X-ray rocking curve is not changed
 after the annealing, the improvement of crystalline quality
 for InN by the annealing is concluded not to be in macro-
 scopic scale but microscopic scale. No improvements are
 found for the samples annealed in the N_2 flow. No data that
 show the chemical oxidation of InN are also found. Further
 investigations will be needed to clarify the mechanism of
 the improvements although the oxygen passivation of do-
 nors such as hydrogen is considered as one of the possible
 causes for the improvements.

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