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

Marked improvements in electrical and optical properties for MOVPE InN annealed at a low temperature (300°C) in O_2 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 E_2 (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 N_2 flow. No data that show the chemical oxidation of InN are also found.

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, $0.07m_0$[1]-$0.14m_0$ [2], and the highest electron drift velocity, 4.2 x 10^7 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 10^{17} cm^{-3} has been realized for MBE InN, while that for MOVPE InN has been still in the range 10^{19}-10^{18} 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 ºC. 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 N_2 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 ºC on (0001) sapphire substrates with a GaN buffer. After the growth, InN samples are annealed at 300ºC for 3-10 h in the air or in the N_2 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 backscattering geometry from the grown surface, with a 458 nm Ar^+ laser.

3 Results and discussion

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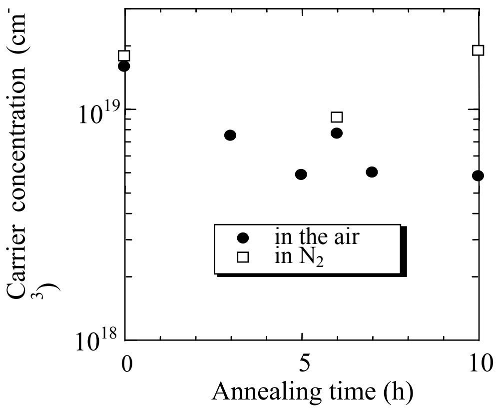

Fig.1. Annealing time dependence of carrier concentration for InN films annealed at 300ºC in the air or N_2 atmosphere.

Figure 1 shows the carrier concentration for InN as a function of post-growth annealing time. By annealing the samples at 300ºC in the air for 5 h, the carrier concentration is reduced from 2 x10^{19} to 5 x10^{18} cm^{-3}. The carrier concentration of 5 x10^{18} cm^{-3} corresponds to the lowest one obtainable for MOVPE InN. No significant change is brought by the annealing in the N_2 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

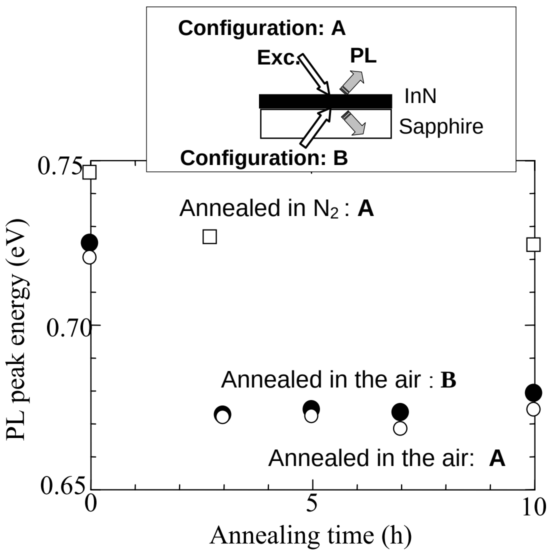

Fig. 2. Annealing time dependence of PL peak energy for InN films annealed at 300ºC in the air or N_2 atmosphere. PL spectrum is measured with the two different configurations, i.e., from the front (configuration A) or the interface side (configuration B) of the films.

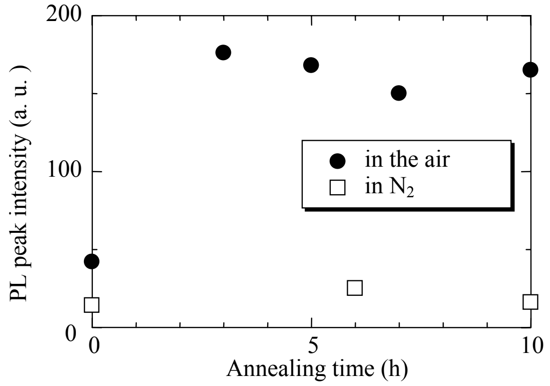

Fig. 3. Annealing time dependence of PL intensity for InN films annealed at 300ºC in the air or N_2 atmosphere.

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 N_2 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

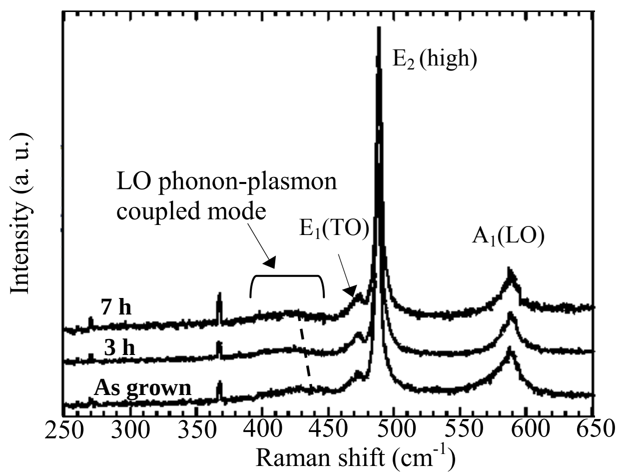

Fig. 4. Raman spectra for as-grown and annealed InN films.

decrease of carrier concentration, supporting the Hall and PL improvements shown in Figs. 1-3. It should be noted that for all the samples, no In_2O_3-related peaks are found. This means that no chemical oxidation occurs in this case. Figure 5 shows the annealing time dependence of the Raman shift and FWHM of the E_2 stress in InN is not changed by the annealing. As seen in Fig. 5, FWHM of the E_2 (high) mode is slightly decreased. (high) mode. No shift of the E_2 (high) is observed for the annealed samples. Therefore, one can see that the residual

Therefore, the crystalline quality of InN film is slightly improved by the annealing in the air. Figure 6 show the annealing time dependence of X-ray rocking curve (XRC) for InN films annealed at 300ºC in the air. It is clear that the XRC-FWHM is not changed by the annealing in the air. Therefore, it is concluded that the improvement of crystalline quality for InN by the annealing is not in macroscopic scale but microscopic scale.

The cause for the improvements of electrical and optical properties found in this study is not clear at present. One of the probable reasons may be the oxygen passivation of donors such as hydrogen incorporated during the growth. Further investigations will be needed to clarify the mechanism of the improvements.

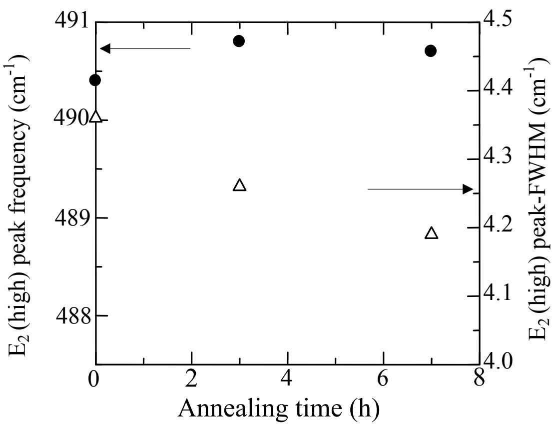

Fig. 5. Annealing time dependence of E_2 (high) peak frequency and FWHM.

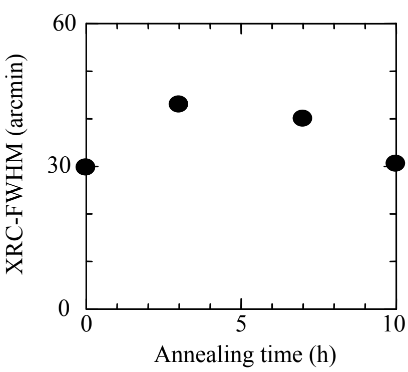

Fig.6. Annealing time dependence of FWHM of (0002) X-ray rocking curve for InN films annealed at 300ºC in the air.

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 $x10^{19}$ to 5 $x10^{18}$ cm^{-3}. In accordance with the carrier reduction, the PL intensity is increased by the factor of about 3 and the PL peak energy is shifted from 0.73 eV for an as-grown sample to 0.67 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 E_2 (high) mode is slightly decreased, indicating the crystalline quality is improved by the annealing. 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 macroscopic 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 donors such as hydrogen is considered as one of the possible causes for the improvements.

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