Nanoscale photoluminescence mapping for MOVPE InN films using scanning near-field optical microscopy (SNOM)

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Abstract

This paper reports for the first time the scanning near-field optical microscopy (SNOM) analysis of MOVPE InN. A near-field PL spectrum and its intensity mapping for MOVPE InN are obtained successfully at room temperature. The near-field PL spectrum has a smaller FWHM and a little higher peak energy compared with the conventional macroscopic PL spectrum. Near-field PL images are used to know the effects of GaN buffer layer on in-plane optical uniformity in MOVPE InN. A large non-uniformity is seen in the image for the sample grown without GaN buffer. Compared with the film grown without buffer, the film grown with a GaN buffer has a better uniformity. Although the use of buffer improves the apparent in-plane uniformity, a fine structure is found in both the PL and topographic images. The fine structure seems to be related to the small grains of InN grown on the GaN buffer composed of small grains.

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1. Introduction

The interest in InN as a highly potential material for high-speed electronic devices has been markedly increased because of the small electron effective mass and the high theoretical maximum electron mobility in InN. Optical devices operating in the wavelength region from ultraviolet to infrared, including a tandem solar cell, can be made in the use of InGaN, InAlN and InGaAlN alloys, since InN has a direct band gap of 0.7 eV. Although significant improvements in the growth of InN films have been attained recently by using molecular beam epitaxy (MBE) or metalorganic vapor-phase epitaxy (MOVPE), films applicable to device fabrication have not yet been grown. Further studies on InN, especially from the standpoints of growth and characterization, are needed to realize InN-based devices. It should be pointed out that most of electrical and optical data obtained for InN films are analyzed based on the assumption that those films are homogeneous. However, there are some evidences, which show inhomogeneities in InN. The ‘surface electron accumulation layer’ reported for MBE samples [1] is one of the typical examples. A large difference between PL peak energy and absorption edge for MOVPE InN [2] compared with that for MBE samples [3] seems to show that MOVPE InN films have a relatively larger inhomogeneity in their properties. Therefore, studies on inhomogeneities in InN are highly required to improve the film quality for device applications. In this work, we employ the scanning near-field optical microscopy (SNOM) to analyze non-uniformity in MOVPE InN films. The study is focused on the comparison of non-uniformity between InN films grown without and with GaN buffer.

2. Experimental

Atmospheric-pressure MOVPE method is used to grow InN films. The films are grown on nitrided (0001) sapphire substrates at 600 °C in the pressure of 800 Torr. A 20 nm thick GaN layer grown at 550 °C is used as a buffer. Samples without buffer are also prepared for the comparison. The carrier concentration in the samples used for the study is $5 \times 10^{18}$ to $3 \times 10^{19}$ cm$^{-3}$. The setup of SNOM system (Type NFS-220FK, JASCO Corp., Japan) is schematically shown in Fig. 1. In the SNOM system, a green laser ($\lambda = 532$ nm, $P = 100$ mW) is used as an excitation source. A scanning probe with an aperture size of 100–500 nm is used to excite the sample surface and collect near-field emission.
(illumination–collection mode). The measurement is made at room temperature. The emission is analyzed using FTIR spectrometer with two different types of InGaAs pin photodiode; One (Detector I; Type 220FK-01, JASCO) has a cut-off wavelength 1600 nm and the other (Detector II; Type G7754-01 (LN-cooled), Hamamatsu Photonics) 2300 nm. The scanning probe is vibrated at its resonance frequency by the piezoelastic ceramic transducers. The photodiode detects vibration amplitude of the probe. With decreasing sample–probe separation, the amplitude of the probe is decreased because shear force interaction between the probe and the sample surface is increased. By monitoring vibration amplitude, the sample–probe separation is kept at a constant value. When we scan the sample while shear force feedback is working, the sample position (height) changes depending on the sample topography. Thus, sample topography can be mapped by monitoring the stage movements at each point of measurement. A conventional macroscopic PL spectrum is also measured at room temperature by using a grating monochromator, a He–Cd laser (λ = 442 nm, P = 300 mW) as an excitation source and an InGaAs pin photodiode (Type G7754-01 (LN-cooled), Hamamatsu Photonics) as a detector. The detector used here is of the same type as the Detector II in the SNOM measurement. All spectra shown in this paper are corrected using the spectral response of each detector. In order to check the effects of wavelength dispersion of the spectrometers (FTIR and grating monochrometer), a macroscopic PL spectrum is also measured using the FTIR system with the Detector II. In this measurement, the fiber probe is not used and, therefore, the excitation light is not focalized into a small size (~500 nm) and a far-field PL is recorded. Fig. 2 shows the comparison of the macroscopic PL spectra measured with the FTIR and with the grating monochrometer. As shown in the figure, no difference is found between the both. Therefore, we can compare the near-field PL measured with the Detector II with the conventional macroscopic PL.

3. Results and discussion

Fig. 3 shows the near-field PL spectra measured with the different detector (Detector I or II). A scanning probe with an aperture size of 500 nm is used in this case. Also shown in Fig. 2 is the macroscopic PL spectrum for the comparison. Even at room temperature a near-field PL spectrum is measured successfully as seen in Fig. 3. The emission mechanism of PL with the peak energy around 0.7 eV is known to be the recombination between degenerate electrons and free holes [4]. Therefore, the PL peak energy is increased with increasing...
carrier (electron) concentration in the samples (the Burstein–Moss shift). The Detector I cannot cover the whole spectrum because of its cut-off wavelength 1600 nm, while the Detector II can cover the whole range of the emission from InN although the signal is much noisy. One can see that the near-field PL spectrum (detected by the Detector II) has a higher peak energy compared with the macroscopic PL spectrum. Since, the SNOM measurement system with the Detector II is confirmed to give a similar PL spectrum as the macroscopic one when the fiber probe is not used to decrease the spot size of the excitation as shown in Fig. 2, the higher PL peak energy in the near-field PL spectrum measured with the Detector II is related to the increase in excitation intensity by decreasing spot size of the excitation. The high excitation density ($\sim 10$ MW/cm$^2$) may cause thermal and/or optical deterioration of a measurement point on the sample surface. The temperature increase may also occur at the measurement point due to the high excitation density though the PL peak shift due to the temperature increase seems to be difficult to detect because of the very small temperature dependence of band gap energy for InN [5].

The effect of band filling to degenerated tail states under the high-density excitation should also be taken into account. In Fig. 3, one can see that the near-field PL measured with the Detector II has a smaller FWHM compared with the macroscopic PL. The reason for this may be related to that the emission is from a limited small area (>500 nmφ) of the sample surface, since InN samples have considerably large in-plane inhomogeneity in quality as shown below. In order to analyze the near-field PL spectra in detail, we need further experiments including SNOM measurements at a low-temperature.

Fig. 4 shows the line scanned near-field PL spectra measured with a 500 nmφ probe at 0.5 μm intervals. In this measurement the Detector I is used. It is clearly seen that the emission intensity is markedly dependent on the position and it changes by more than factor 2 when the measurement point is shifted by only 0.5 μm. Fig. 5 shows the near-field PL intensity images (10 μm×10 μm size) for InN films grown with and without GaN buffer. For this measurement, a 500 nmφ probe and the Detector I are used and peak-area intensity of spectra is plotted with 0.5 μm intervals. The topography image is recorded simultaneously and the results are also shown in Fig. 5. One can see that the feature of the PL image is totally different between both the samples. The sample grown without buffer has a considerable inhomogeneity in PL intensity and some areas of the film show a very low PL emission. Such a low PL intensity is due to the existence of low quality grains. This may be related to the fact that the nucleation of InN on a sapphire is not easy and is very inhomogeneous [6]. Such a large inhomogeneity seen in the PL image is not found in the topography image. On the contrary, the sample grown with a GaN buffer shows a relatively uniform PL image and both the PL and topography images are very similar each other. Thus, the use of the buffer is found to improve the apparent in-plane uniformity. However, a fine structure is seen in the both images. The fine structure indicates that the film is composed of small grains and the grain growth is suppressed. This is due to that both the nucleation and grain growth of InN are

![Fig. 4. Line-scanned near-field PL spectra for InN measured at 0.5 μm intervals with a 500 nmφ probe and the Detector I.](image1)

![Fig. 5. Near-field PL intensity images and surface topographies (10 μm×10 μm size) plotted with 0.5 μm intervals for InN films grown with and without GaN buffer. A scanning probe with an aperture size of 500 nmφ is used.](image2)
governed by the small grains of the GaN buffer [7]. Thus, the use of a GaN buffer is not a perfect solution for the improvement of MOVPE InN although it improves the apparent in-plane uniformity. Some procedures are required to enhance grain growth for the improvement of MOVPE InN quality.

4. Conclusion

In order to get information about in-plain uniformity in MOVPE InN films, the scanning near-field optical microscopy (SNOM) analysis has been applied for the first time. A near-field PL spectrum is obtained at room temperature. It is found that the near-field PL spectrum has a smaller FWHM and a little higher peak energy compared with the macroscopic PL spectrum. Intensity mapping image of near-field PL spectrum and surface topography are obtained simultaneously and compared each other. A large non-uniformity in the near-field PL image is observed for the sample grown without buffer. Such a large non-uniformity is not observed in the topography. Compared with films grown without buffer, those grown with a GaN buffer have a better uniformity in near-field PL image. The fine structures observed in both the PL and topography images for the film grown with GaN buffer indicate that the grain growth of InN is markedly suppressed, although the apparent in-plane uniformity is improved by the use of a GaN buffer. This is due to that both the nucleation and grain growth of InN are governed by the small grains of the GaN buffer.

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