Extended Abstracts of the 2002 International Conference on Solid State Devices and Materials, Nagoya, 2002, pp. 526-527

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Experimental Consideration of Optical Band-Gap Energy of Wurtzite InN

Takashi Matsuoka, Masashi Nakao[1], Hiroshi Okamoto[1], Hiroshi Harima[2] and Eiji Kurimoto[3]

NTT Basic Research Laboratories
3-1, Morinosato Wakamiya, Atsugi-shi, Kanagawa 243-0198, Japan
Phone: +81-46-240-3122 Fax: +81-46-240-4729 E-mail: matsuoka@will.brl.ntt.co.jp
[1]NTT Photonics Laboratories
3-1, Morinosato Wakamiya, Atsugi-shi, Kanagawa 243-0198, Japan
[2]Kyoto Institute of Technology, Dept. of Electronics and Information Science
Matsugasaki, Sakyo-ku, Kyoto 606-8585, Japan
[3]Osaka University, Dept. of Applied Physics
2-1 Yamadagaoka, Suita-shi, Osaka 565-0871, Japan

1. Introduction

GaN, AlN, and InN have recently attracted great interest as materials for bright light-emitters and switching devices. Among them, InN remains the most mysterious compound, due mainly to difficulty in growing high-quality crystals because of the extremely high equilibrium vapor pressure of nitrogen [1]. A typical issue is the fundamental band gap E_g. Early absorption studies on poly-crystalline films grown by discharge[2], and sputtering studies [3,4,5] reported E_g=1.8-2.0 eV at room temperature. All of those samples showed no corresponding band-edge photoluminescence (PL). It is quite recently that band-edge emission was observed below 200 K in InN layers grown on Si by molecular beam epitaxy (MBE) [6]. The emission peaks observed at 1.81-2.08 eV were interpreted as a mixture of hexagonal and cubic InN components. To our understanding, however, none of these results showed conclusive evidence of E_g~1.9 eV in high-quality single crystals of wurtzite InN.

We previously reported [1], based on a series of absorption experiments on high-quality wurtzite $In_xGa_{1-x}N$ films, that E_g decreased monotonically from 3.4 eV (x=0) to 2.1 eV (x=0.42), and pointed out that InN single crystals would have much smaller band gap than commonly believed (~1.9 eV) based on early experiments. Actually, E_g~0.9 eV has been very recently reported using high-quality InN films grown by MBE in experiments on PL and PL-excitation at low temperature and absorption [7].

Here, we present the experimental evidence that further supports the existence of a similar band gap in high-quality InN films grown by metalorganic vapor phase epitaxy (MOVPE). At room temperature, we have observed strong band-edge PL peaks at 0.76 eV as well as an absorption edge at 0.7-1.0 eV.

2. Experiments

The samples were grown by a two-step MOVPE at ambient pressure with a structure of InN (0.1-0.2 m thick)/GaN (1.6 m thick)/sapphire (0001). The growth temperature was 1010.C for GaN and 500.C for InN. TMI (trimethylindium), TMG (trimethylgallium) and ammonia were the source gases, and the flow ratio of ammonia to TMI (V/III ratio) was 1.6×10^5 to 6.6×10^5 during the InN growth. At this growth temperature, ammonia decomposes by less than 0.1%. As a carrier gas, nitrogen was used instead of hydrogen to promote ammonia decomposition. Following our established procedure [1], single-crystal growth of a (0001)-oriented hexagonal InN layer was carefully confirmed by reflection high-energy-electron diffraction (RHEED) and ω-2θ scan of x-ray diffraction (XRD). Surface morphology was observed by optical microscopy. A confocal Raman microprobe was employed to observe the crystallinity of the InN layer. PL and absorption experiments were carried out to examine the fundamental band gap.

3. Results and Discussion

The samples were brownish in color. Observation with an optical microscope in interference derivative mode showed excellent surface morphology with no metal-indium droplets.

Figure 1 shows the dependence of the spectra of XRD in the ω-2θ scan mode on the V/III ratio during the InN growth.

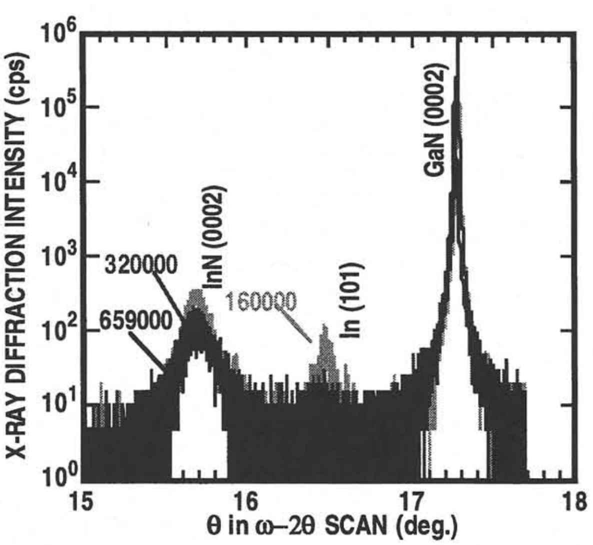

Fig. 1 Dependence of the spectra of XRD in the ω-2θ scan mode on the V/III ratio during the InN growth.

At the V/III ratio of 6.6×10^5, the In (101) signal, which is often observed when indium segregated, was below the detection limit. XRD showed that the samples did not include the cubic phase component. Only InN grown using this high V/III ratio is described below.

The features of Raman spectra resemble very well those of the highest-quality (0001)-oriented InN films ever reported [8]. Comparison of LO-phonon-plasmon-coupled mode with a previous report [8] suggests that the InN layer has a high carrier concentration of $n = 5\text{-}10\times10^{19}$ cm^{-3}. On the contrary, the observation of the nearly uncoupled A_1 (LO)-phonon mode indicates the existence of a fairly low-carrier-density region. Such co-existence of low- and high-carrier-density regions is commonly observed in recent high-quality InN epitaxial layers, and remains a kind of mystery [8]. Highly oriented characteristics were confirmed also by polarized Raman scattering.

A typical result of the absorption experiment is shown in Fig. 2. Extrapolation of the linear region I to the horizontal axis gives the band gap of the underlying GaN layer. On the contrary, region II gives E_g = 0.7–1.0 eV, which should be ascribed to the InN layer. The latter value was not rigorously determined because of thin layer-thickness of InN and the strong absorption tail of region I from the thick GaN layer.

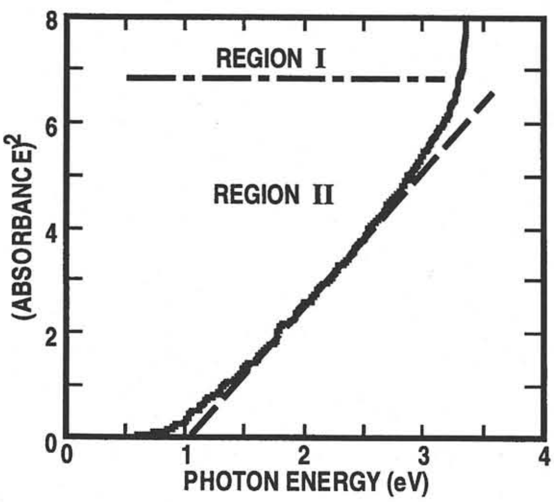

Fig. 2 Absorbance squared versus photon energy.

Figure 3 shows PL spectra measured at room temperature. Excitation and detection in region A and B was carried out using a YAG laser and an InGaAs-photodetector with a cut-off wavelength of 2.05 m, and the second harmonic wave (375 nm) of a Ti-sapphire laser with the power of 0.6 MW/cm^2 and Si-photodetector, respectively. The PL peak was observed at 0.76 eV independently on the excitation power. Even under high-power excitation, no PL was observed near the previous band gap energy (~1.9 eV). By comparing Figs. 2 and 3, we consider that Fig. 3 shows a near-band-edge emission. To conclude, our results strongly suggest that optical band gap of InN should be 0.8-1.0 eV.

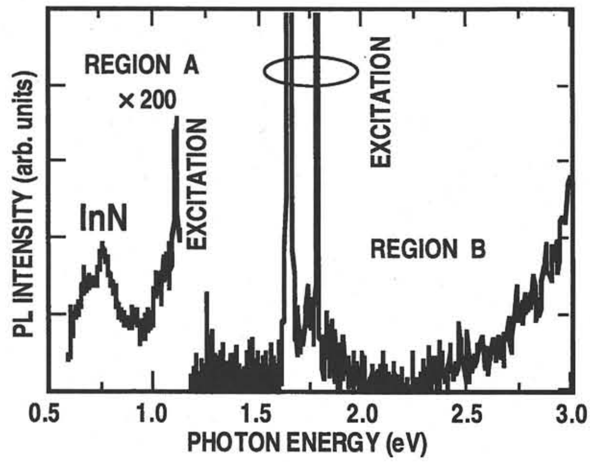

Fig. 3 PL characteristics at room temperature.

4. Conclusions

Wurtzite InN films grown on a thick GaN layer by MOVPE showed well-oriented single crystals with high crystallinity, as confirmed by Raman scattering, XRD, and RHEED. An absorption edge at 0.7-1.0 eV and strong PL signals at 0.76 eV were observed at room temperature. These results suggest that the optical band gap of InN should be 0.7-1.0 eV. The discrepancy from previous data could be due to the difference in crystallinity.

Acknowledgments

The authors thank Miss Emi Hagiwara for the X-ray diffraction measurement. They also thank Drs. Takaaki Mukai and Naoki Kobayashi for the opportunity to do this work

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