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Recombination mechanism of photoluminescence in InN epilayers

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Abstract

We report an investigation of the recombination mechanism for photoluminescence (PL) in InN epilayers grown by molecular beam epitaxy and metal-organic chemical vapor deposition with a wide range of free electron concentrations from 3.5×10^{17}–5×10^{19} cm^{-3}. We found that the PL spectra are strongly blueshifted with increasing excitation intensity. For all the samples studied, the exponent of the relationship between the integrated PL intensity and the excitation intensity is very close to unity and independent of the temperature. By assuming Gaussian fluctuations of the random impurity potential, calculation based on the 'free-to-bound' recombination model can be used to interpret our results very well and it correctly reproduces the development of the total PL peak shift as a function of carrier concentration. It is concluded that the PL transition mechanism in InN epifilms can be characterized as the recombination of free electrons in the conduction band to nonequilibrium holes in the valence band tail.

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1. Introduction

InN, the least studied Group-III nitride, has attracted intense interests because of its distinctive physical properties [1–3]. Early reports using infrared absorption technology have shown that the band gap energy of InN is 1.8–2.0 eV [4] Recently, numerous studies have observed instead a strong PL emission at around 0.7–0.8 eV, leading to the revision of the fundamental band gap of InN from 1.8–2.0 to 0.7–0.8 eV [5–7]. This newly discovered discrepancy has spurred numerous attempts to incorporate InN in various optoelectronic devices. For instance, the entire visible spectrum can now be spanned by a single material system; therefore, InN and its alloys have also emerged as promising materials for optoelectronic devices such as light-emitting diodes and solar cells. In addition, the band gap energy of InN makes it suitable for application in 1.55 μm-telecommunications.

In order to effect the application in optoelectronic devices, it is essential to have a detailed understanding of the optical properties of InN thin films. Recently, Arnaudov et al. have made an important breakthrough and they reported that the mechanism responsible for the emission at 0.7–0.8 eV can be very well described a model of 'free-to-bound' radiative recombination of degenerate electrons from the conduction band with non-equilibrium holes located in the valence band tails [8]. This model, in principle, resolves the question about the recombination mechanisms in the near band edge region. However, recent low temperature absorption measurements led Shubina et al. to propose that the IR emission at around 0.7 eV may be due to the analogous Mie scattering produced by In clusters [9]. Very recently, Guo et al. have observed a visible luminescence from InN thin films grown by reactive radio frequency magnetron sputtering and inferred that the emission at 0.7–0.8 eV could be due to In clusters or oxygen-related defects [10]. In addition, Butcher et al., Alexandrov et al. and Intartaglia et al. have also shown that the emissions around 0.7–1.0 eV were consistent with the presence of 0.7 eV deep level trap and/or the recombination of electron–hole pairs or excitons to extrinsic defects or topological defects [11–14]. Thus, the luminescence mechanism(s) of InN are still discordant. As a consequence, further investigation of the optical properties of this material has to be undertaken to clarify the inconsistency.

In this paper, we present a study of the PL recombination mechanism of InN epilayers with a wide range of free electron

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concentrations ($n=3.5\times10^{17}$ cm^{-3} to $n=5\times10^{19}$ cm^{-3}) grown by MBE and MOCVD. We found that the PL spectra are blueshifted with increasing excitation intensity. The exponent of the relationship between the integrated PL intensity and the excitation intensity does not depend on temperature and has a value close to unity. By assuming Gaussian fluctuations induced by random impurity potential, calculation based on the 'free-to-bound' recombination model can be used to interpret our results very well. In addition, the development of the total PL peak shift (defined as the difference of the PL peak energy obtained from the highest and the lowest excitation intensity) as a function of free electron concentration can also be well characterized by the same model. We thus conclude that the 'free-to-bound' recombination model is the underlying mechanism responsible for the PL recombination in InN epifilms.

2. Experiment

InN films were grown on (0001) sapphire (sample set I and III) and (111) silicon (sample set II) substrates with AlN or GaN (sample set I and III) and β-Si_3N_4/AlN (0001) (sample set II) buffer layers by MBE (sample set I and II) and MOCVD (sample set III). The thickness of the buffer layer ranges from 70 to 300 nm. The InN layer thickness is between 250 nm and 7.5 μm, which implies that all the InN films were fully relaxed. Although most of the samples were not intentionally doped, free-electron concentrations ranging from 3.5×10^{17} to 5.5×10^{18} cm^{-3} (sample set I), 1×10^{19} cm^{-3} (sample set II and III) have been found in these samples by Hall effect measurements. Higher free-electron concentrations were achieved by doping InN with Si. The free-electron concentration of these doped samples varies between 1.0×10^{19} and 5.0×10^{19} cm^{-3}. The details of the growth process have been published elsewhere [15–17]. X-ray diffraction studies show that these InN epitaxial layers have a high quality and a wurtzite structure with their *c*-axis perpendicular to the substrate surface. Hall mobilities range from several hundred to 2050 cm^2/V s.

The PL spectra were dispersed by a Spectra Pro 275i monochromator, and detected by an EOS extended InGaAs detector which has a cut off wavelength of 2.4 μm. A semiconductor diode laser working at 808 nm was used as the excitation source. The excitation intensity was varied from 3.9×10^{-4} to 3.9 W/cm^2. All the measurements were carried out at 20 K.

3. Results and discussion

Typical PL spectra observed under different excitation intensities are shown in Fig. 1(a)–(d). Clearly, the PL spectrum with higher free electron concentration peaked at a higher energy position. Such behavior can be well described by the combination of the Kane's model, Burstein–Moss effect, band tailing effect, and band renormalization effect as shown in the previous reports [18,19]. The analysis of the influence of carrier density on PL spectra is, therefore, not the goal of this paper. Instead, we would like to focus on the origin of the

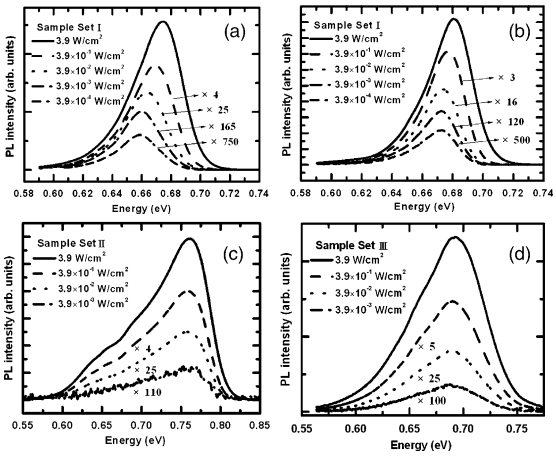

Fig. 1. Photoluminescence spectra of InN samples under different excitation intensities. (a) Sample set I, $n=5.7\times10^{17}$ cm^{-3}, (b) sample set I, $n=1.425\times10^{18}$ cm^{-3}, (c) sample set II, $n=9\times10^{18}$ cm^{-3}, (d) sample set III, $n=1\times10^{19}$ cm^{-3}.

recombination mechanism of this material, which is still inconclusive at the present stage. Second, the PL peak energies shift to higher energy rapidly with increasing excitation intensity for the low degenerate sample as shown in Fig. 1(a). This result strongly indicates that the recombination mechanism at issue is not 'direct band to band' transition as reported previously [16]. Comparing the PL spectra for the higher degenerate samples shown in Fig. 1(b)–(d) to the previous report, it is clear that the discrepancy may be due to the fact that the residual carrier concentration of those measured samples were too high for the determination of the exact recombination mechanism for pure InN films [16].

Fig. 2 shows the integrated PL intensity on the logarithmic scale as a function of excitation intensity for sample sets I, II, and III. The relationship can be expressed as:

$$I_{PL} = \eta I_{EX}^{\alpha} \tag{1}$$

where η is a coefficient which depends on temperature, I_{PL} is the integrated PL intensity and I_{EX} is the intensity of the excited source [20,21]. It is well known that the exponent, α, strongly depends on the radiative recombination mechanism

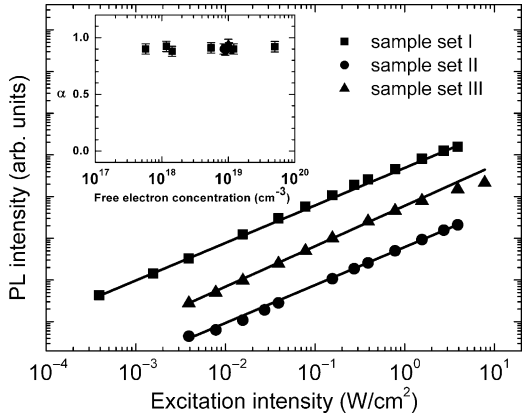

Fig. 2. Integrated photoluminescence intensity as a function of excitation intensity. The theoretical fitting using Eq. (1) is shown as the solid lines. Inset shows α as a function of free electron concentration for all studied samples.