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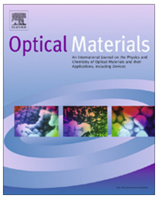

Luminescence studies in $In_xGa_{1-x}N$ epitaxial layers with different indium contents

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ABSTRACT

The optical properties of $In_xGa_{1-x}N$ epitaxial layers ($x = 0.02$, 0.04, 0.11, 0.15, 0.30 and 0.33) grown by metalorganic chemical vapor deposition (MOCVD) have been investigated by temperature-dependent photoluminescence (PL) measurement. The surface morphologies of InGaN samples are studied by scanning electron microscopy (SEM) images. The PL feature at 12 K has shown an increase in full-width at half-maximum (FWHM) with increasing In content. An anomalous S-shaped temperature dependence of the PL peak energy exhibited by InGaN films with higher In content enabled the evaluation of the exciton localization energy. The broadened FWHM and S-shaped emission shift are attributed to larger compositional fluctuation due to compositional inhomogeneity of In. Additionally, the luminescence mechanism relating to the phase separation has to be considered for the much larger FWHM value and the pronounced S-shaped behavior for the InGaN samples with In content of 0.30 and 0.33.

1. Introduction

Recently, devices based on III-nitride compound semiconductors which include GaN, InGaN, and AlGaN materials have been rapidly and widely developed for highly efficient laser diodes, high brightness light emitting diodes (LEDs), photodetectors, etc. [1–4]. Among the III-nitride compound semiconductors, the direct band-gap InGaN-based semiconductors have attracted much attention for versatile optoelectronic device application because the energy gap can be tuned in a rather wide range from 3.4 eV for GaN in the ultraviolet (UV) region to 0.7 eV for InN in the infrared (IR) region [5,6]. However, the big difference in the covalent radii of In and Ga would result in inhomogeneous In distribution in InGaN films with higher In content, affecting the optical properties of the InGaN sample [6].

Up to date, several luminescence properties have been discussed for InGaN alloy semiconductor. It has been reported that the compositional fluctuation is one of the most important subjects with respect to the optical emission in InGaN alloys [7–10]. The compositional fluctuation has been known to be responsible for the broadening in the full-width at half-maximum (FWHM) of the luminescence feature and the observed ‘S-shaped’ temperature dependence of luminescence peak energy (red–blue–redshift with increasing temperature) relating to the exciton localization effect [11–14]. However, to our knowledge, detailed discussion concerning the relationship between the variation in FWHM and S-shaped emission shift for the InGaN with different In contents has not been well established.

In this work, we have investigated the luminescence properties of $In_xGa_{1-x}N$ grown by metalorganic chemical vapor deposition (MOCVD), as a function of x ($x = 0.02$, 0.04, 0.11, 0.15, 0.30 and 0.33). The temperature-dependent photoluminescence (PL) were measured to determine the dependence of the band-gap energy of InGaN on the In composition. The FWHM values and temperature-dependent peak energy of PL spectra taking into account the exciton localization effect are analyzed and discussed.

2. Experimental procedures

The $In_xGa_{1-x}N$ films investigated were grown by MOCVD system. On (1000) sapphire, a thin, about 30-nm thick, GaN buffer was first grown at low temperature (LT) at 530 °C, followed by approximately 1 μm thick GaN deposited at 1030 °C and finally followed by a nearly 100 nm thick InGaN layer grown at 700 °C. The composition values of these samples were determined by high resolution X-ray diffraction. Growth details of the samples can be found in Refs. [15,16]. The surface morphologies of InGaN samples observed by scanning electron microscopy (SEM) images were taken on a JEOL-JSM7001F. Temperature-dependent photolumines-

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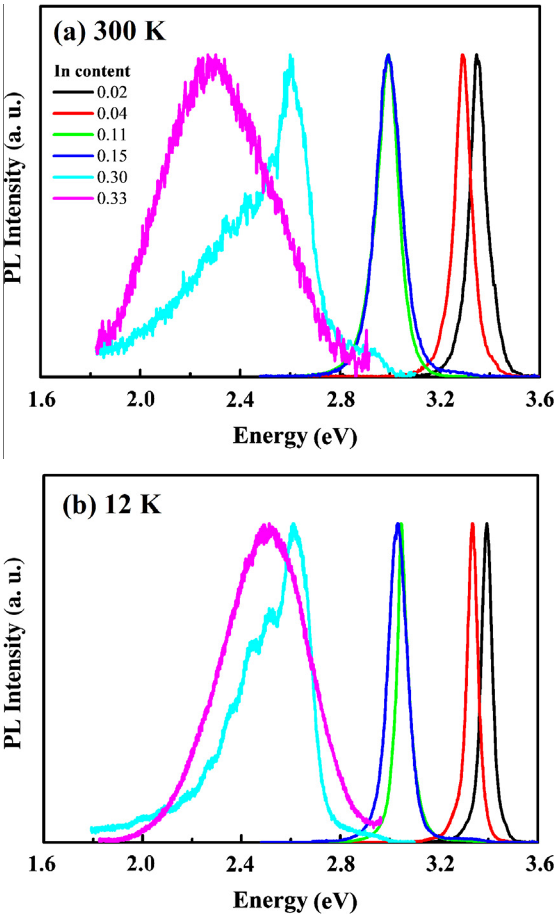

Fig. 1. PL spectra for the $In_xGa_{1-x}N$ films with different In contents measured at: (a) 300 K, and (b) 12 K.

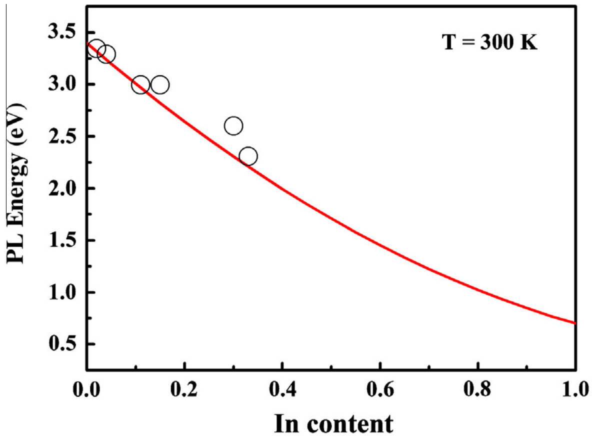

Fig. 2. Band-gap of the $In_xGa_{1-x}N$ films as a function of In content. The solid line is the fitted curve from Eq. (1).

cence (PL) measurements were conducted under the excitation with a 5 mW/cm^2 of a microchip laser (266 nm). The luminescence was collected using a spectrometer (Zolix omni-λ 500) with a 1200 grooves/mm grating and detected using a GaAs photomultiplier tube. The PL signal obtained from the photomultiplier was analyzed using lock-in technique and recorded in a computer. In addition, Janis Research Model CCS-150 and LakeShore Model 321 temperature controller were used to measure the PL spectrum as a function of temperature. The computational parts for the theoretical calculation in the present work are facilitated by commercially available Mathcad software.

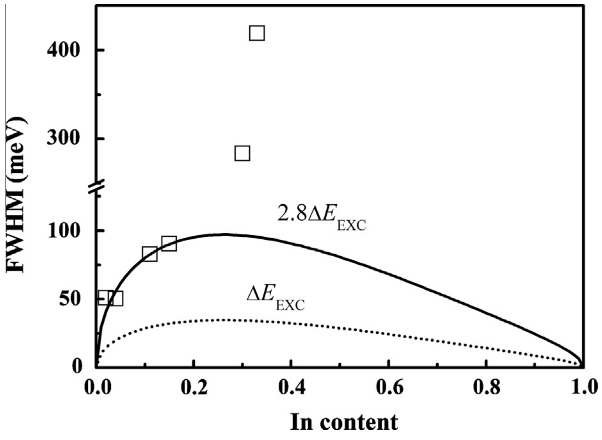

Fig. 3. The variation of the 12 K PL FWHM for the $In_xGa_{1-x}N$ films with different In contents.

3. Results and discussion

Fig. 1a and b shows the normalized PL spectra of $In_xGa_{1-x}N$ films with different In contents at 300 and 12 K, respectively. Here, the normalization is with respect to the peak intensity of the PL feature. In Fig. 1a, peak energies of the PL signals measured at 300 K are estimated to be 3.35, 3.29, 2.99, 2.99, 2.59, 2.31 eV for the sample with $x = 0.02$, 0.04, 0.11, 0.15, 0.30 and 0.33, respectively. In the present analysis, we assume that the band-gap of InGaN can be directly obtained from the maximum of the PL peaks. The obtained band-gap of the InGaN samples as a function of In content are then plotted in Fig. 2.

It has been known that the band-gap values for a ternary compound $A_xB_{1-x}C$ do not vary linearly with the band-gap values of the two binary ends, $E_{g,AC}$ and $E_{g,BC}$ The variation for the band-gap of $In_xGa_{1-x}N$ is more appropriately described by [5,17,18]:

$$E_{g,\mathrm{InGaN}}(x) = xE_{g,\mathrm{InN}} + (1-x)E_{g,\mathrm{GaN}} - bx(1-x) \tag{1}$$

where b is the bowing parameter and has units of energy. In Fig. 2, the red[1] solid line is the theoretical line least squared fit to Eq. (1) using the reported values of $E_{g,\mathrm{InN}} = 0.7$ eV, $E_{g,\mathrm{GaN}} = 3.4$ eV, with the bowing parameter b estimated to be 1.36 eV. The value is very similar to the obtained value of 1.43 for In-rich InGaN [17], and is in agreement with the recent calculations predicting a value of 1.36 eV for the whole composition range [18]. However, a slight deviation from Eq. (1) is observed for InGaN samples with $x > 0.11$, which can be attributed to a larger degree of compositional inhomogeneity in In distribution for the In-rich samples [17,18].

From the 12 K PL spectra (Fig. 1b), it can be observed that the PL shape tends to become asymmetric with a low energy tail and increased FWHM with increasing In content >0.11. The relationship between the degree of compositional fluctuation and the broadened PL features as a function of In content is analyzed as follows. The estimated FWHM as a function of In content are plotted as opened squares in Fig. 3, and the theoretical variation in the FWHM, ΔE_{EXC}, of the PL peaks can be described by [19,20].

$$\Delta E_{\mathrm{EXC}} = 2.36\sigma_E \tag{2}$$

where σ_E is the standard deviation of the band gap energy given by [20].

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[1] For interpretation of color in Figs. 2 and 4, the reader is referred to the web version of this article.