Observation of large spectral blue-shift in photoluminescence spectra of Mg-doped gallium nitride Nanorods


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1. Introduction

The gallium nitride (GaN) nanostructures including quantum dots and wires have attracted considerable interest in recent years for potential application in novel light emitting devices such as lasers and light emitting diodes. Recently, Johnson et al [1] demonstrated optical pumped single nanowire laser emission from the synthesized GaN nanowires, and S. Tanaka et al [2] reported stimulated emission from self-assembled GaN quantum dots. The photo-luminescence (PL) emission wavelengths of these nanorods were shown to either have the same band-gap energy as the bulk GaN[3], or spectral blue shift depending on the size of nanorod with the latter attributed to the quantum confined effect.[4] On the other hand, red-shift of PL spectrum from the synthesized GaN nanowires had also been observed and was attributed to the biaxial compressive strain effect.[5] As for the self-assembled GaN quantum dots, study of the PL emission spectra indicated the peak energy generally depends on the balance between quantum confinement and strain related effects such as strain induced piezoelectric field.[6] Recently we have reported the fabrication of nanorods from the Mg-doped GaN thin film by inductively coupled plasma reactive ion etching (ICP-RIE).[7] In this paper we report the PL emission characteristics of Mg-doped GaN nanorods and show that the observation of large spectral blue shift of PL peak energy with respect to GaN film is due to the Coulomb interaction between the distant donor-acceptor pair transition.

2. Experiments

The nanorods were fabricated from the Mg-doped GaN film by ICP-RIE etching technique. [7] A non-nanorod region of covered by nickel serves as a reference to compare the PL emission spectrum with respect to the nanorod region. The typical scanning electron microscope (SEM) images of the Mg-doped GaN nanorods are shown in Fig. 1. Figure 1 is the top view image of the Mg-doped GaN nanorods, and the density of the nanorods was estimated to be about $3 \times 10^{10}$ cm$^{-2}$. A micro PL investigation was chosen to avoid the spatial variation in the epi-wafer and the inhomogeneous broadening induced by size distribution of nanorods. The sample was placed on a PZT controlled stage with a resolution of 2 nm and excited by a 325 nm He-Cd laser through a 100X objective. The focused spot size on the sample was about 1µm in diameter which corresponds to about three hundreds illuminated nanorods. The PL emission from the sample was collected by the same 100X objective and fed to a 0.32 m spectrometer with a spectral resolution of 0.1 nm and a cooled UV-enhanced CCD.

3. Results and Discussion

Figure 2 shows the typical micro PL spectra obtained from the nanorod region and the non-nanorod region under laser excitation intensity of about 32 kW/cm$^2$. The PL spectra from both regions shows a typical donor-acceptor-pair emission of Mg-doped GaN with an emission at 3.0 eV similar to those reported by others[8-11] and emission peak energy of the nanorod region shows a large blue shift of 64 meV from that of the non-nanorod region. The periodic fluctuation in PL spectrum of the non-nanorod region is caused by Fabry-Perot interference of the epitaxial Mg-doped GaN film. We also have fabricated the undoped GaN nanorods. However, no blue shift was observed at the band edge emission. The PL emission peak energy for both nanorod region and non-nanorod region increase with the excitation intensity as shown in Fig. 3. The PL peak energy increases were 125 meV and 67 meV for nanorod region and non-nanorod region respectively as the excitation intensity varies from 12kW/cm$^2$ to 56kW/cm$^2$. The corresponding spectral shift between nanorods peak and the non-nanorods peak ranging from 8 meV to 67 meV within the excitation intensity range indicates stronger power dependence of the Mg-doped GaN nanorods emission. Previously the PL spectral of Mg-doped GaN films were reported with the blue shift dependent on the excitation intensity [10-11] and the blue shift was attributed to the Coulomb interaction of distant donor-acceptor pair transition. The effect of a distant DAP Coulomb interaction on the DAP transition energy can be expressed by the following equation:[12]

$$h\nu = E_g - (E_A + E_D) + \frac{e^2}{4\pi\varepsilon_0 R_{DA}} - mh\nu_{LO}$$

where $E_g$ is the band gap of GaN, $E_A$, $E_D$ are the donor and acceptor binding energies, respectively, $R_{DA}$ is the mean distance between the involved donor and acceptor, the third term is the Coulomb interaction term which depends on $R_{DA}$ and the last term describes the LO phonon replica.

From the equation we could expect an increase in the transition energy when the average distance $R_{DA}$ of the Coulomb interaction decreases. This condition could occur as the excitation intensity increases causing an increase in
the occupied donor and acceptor centers and results in the shortening of the average distance $R_{DA}$. Our data for the non-nanorod region shows the emission peak energy increase is also 99 meV per decade power which is comparable to the 90 meV per decade power increase reported for the films. [10] Although the mechanisms responsible for larger spectral blue shift of nanorods are not fully understood yet, the Coulomb interaction should be the contributing factor.

4. Conclusion
In summary, we observed large blue-shift of photoluminescence peak in Mg-doped GaN nanorods using micro-PL. The blue shift PL spectrum of the nanorods show typical DAP transition at 3.0 eV band. One of the contributing factors for the large and power dependent blue shift in the PL emission energy could be the Coulomb interaction of the distant DAP in the nanorods.

References