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Time-resolved photoluminescence spectroscopy of InGaN multiple quantum wells

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We have fabricated by metal organic chemical vapor deposition (MOCVD) $In_{0.13}Ga_{0.87}N/GaN$ multiple quantum well (MQW) with thickness as thin as 10 Å and barriers also of th same width on (0001) sapphire substrate. We have investigated this thin MQW by steady-state and time-resolved photoluminescence (PL) in picosecond time scale in a wide temperature range from 10 to 290 K. In the PL at 10 K, we observed a broad peak at 3.134 eV which was attributed to the quantum well emission of InGaN. The full width at half maximum (FWHM) of this peak was 129 meV at 10 K and its broadening at low temperatures was considered to be due to compositional fluctuations and interfacial disorder in the alloy. The narrow width of the quantum well was mainly responsible for the broadening of the emission linewidth. We also observed an intense and sharp peak at 3.471 eV of GaN barrier. From the temperature dependent PL measurements, the activation energy of the InGaN quantum well emision peak was estimated to be 69 meV. The lifetime of the quantum well emission was found to be 720 ps at 10 K, which was explained in terms of the exciton localization arising from potential fluctuations.

Wide bandgap nitride materials have received much attention especially in the development of emitting devices in UV region [1]. the InN has a bandgap of 1.9 eV and GaN has a bandgap of 3.2 eV. Therefore, the ternary compound $In_xGa_{1-x}N$ is suitable for fabricating new hotonic and optoelectronic devices that emit in a wide spectral range [2-6]. Blue lasers and light emitting diodes (LEDs) have long been recognized as important ingredients in digital video disk (DVD) and high density optical memory devices. Recently Nakamura and co-workers [2,3] have attained a major breakthrough in this area by demonstrating continuous wave (cw) lasing in these materials. These lasers basically consist of $In_xGa_{1-x}N$ as an active layer and either GaN or $In_xGa_{1-x}N$ as a barrier in single quantum wells (SQW) or MQWs. Though these devices are commercially available, there is still much room to be improved in the overall performance. We have carried out systematic studies GaN and related materials to examine the optical phenomena and their relevance to the device performance [7-11]. In our recent reprot [7] we have addressed the issues of exciton-phonon interactions, exciton binding energy and their importance in the realization of room temperature semiconductor lasers based on GaN. The major challenge in fabrication GaN based lasers is to lower the threshold current required for laser action in these materials. The other concern is the inherent compositional inhomogeneity in InGaN and its influence on the lasing performance.

In this letter we report the results of our steady-state and time-resolved spectroscopic measurements on ultra-thin InGaN quantum well with well width of as low as 10 Å. Although there have been hitherto numerous investigations on the optical properties of InGaN quantum wells and epilayers [12-21], they are generally restricted to quantum wells of wider width of several nm. Thus, in the present investigation we focused on the strong confinement effects on the optical properties of MQWs.

The MQWs investigated in this work were grown on (0001) oriented sapphire substrate by MOCVD technique. Before growing the quantum well structure we grew a GaN buffer layer at low temperature on the top of the sapphire substrate. The In composition in InGaN quantum well is 13% and the barrier is GaN. The quantum well width is 10 Å and the barrier also has the same width. The period of the MQW is five. A He-Cd laser (325 nm) was employed as an escitation source for PL measurements. A McPherson 1-m monochromator, a photomultiplier (Hamamatsu) and a lock-in Amplifier (Princeton Applied Research) were employed for PL signal detection. The samples were mounted on a cold finger of a closed-cycle helium cryostat and the temperature was varied in the range of 10-300 K. For time-resolved PL experiments, the time correlated single photon counting (TCSPC) method was employed. As and excitation source, a synchronously pumped picosecond dye laser having about 2 ps pulse width at 3.8 MHz repetition rate was employed. The dye laser beam was frequency doubled using a -barium borate (BBO) non-linear optical crystal. The UV light thus generated was used to excite the sample. Our TCSPC system has a time resolution of ~20 ps after deconvolution.

Fig. 1 shows a series of cw PL spectra of InGaN multiple quantum well under various temperatures. In order to obtain the peak positions accurately, we have reconstructed the PL spectrum by using a Lorentzian line-shape function. By this method we have determined the peak positions and intensities of various transitions. The broad peak at 3.134 eV was interpreted as due to InGaN quantum well and the narrow peak at 3.471 eV as due to GaN barrier. The compositional dependence of PL peak position in strained $In_xGa_{1-x}N$ epitaxial layers was reported by Takeuchi *et al* [12]. They observed the PL peak at ~2.85 eV for In composition of 0.13. Thus, the large blue shift observed here is a direct manifestation of strong quantum confinement effect in ultra-thin quantum wells. At 10 K the PL linewidth, contributed by inhomogeneous broadening, was found to be 129 meV. Such large inhomogeneous broadening is believed to arise from the inherent compositional fluctuation in InGaN temary system [21]. The solid phase immiscibility originates from the large difference in interatomic spacing between GaN and InN [22]. However, the PL linewidth in our sample is broader than those reported by others. For instance, Narukawa *et al.* [17] have reported a linewidth of 80 meV for $In_{0.20}Ga_{0.80}N$-$In_{0.05}Ga_{0.95}N$ MQWS with a thickness of 2.5 nm. The larger PL linewidths in our sample can be understood by considering the effects in the limit of strong quantum confinement. In this situation there are two main contributions. One is the interface roughness scattering [23,24] due to well width fluctuation and the other is the increased contribution from the enhanced exciton-phonon interactions in very narrow quantum wells [13-15] This argument is also supported by the time-resolved results see the below.

Fig. 2 shows the temperature dependent cw PL plot of InGaN quantum well from which the activation energy was estimated to be 69 meV. In our previous investigations [7,8] we have found that the free

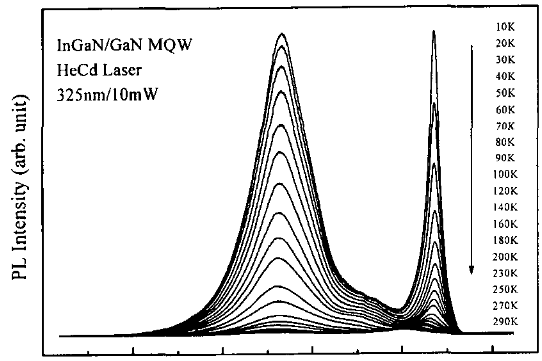

Fig. 1. Photoluminescence spectra of InGaN multiple quantum well under various temperatures. The broad peak at 3.134 eV is due to the InGaN quantum well and the narrow peak at 3.471 eV is due to GaN barrier at 10 K.

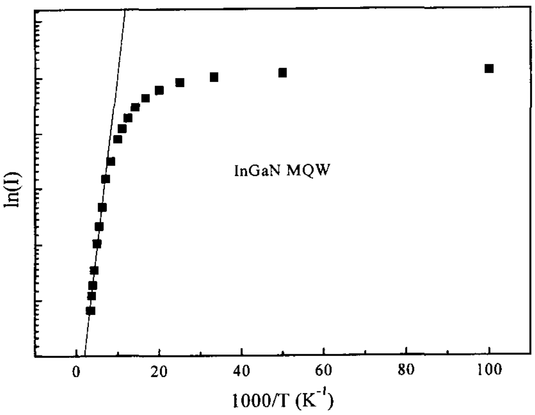

Fig. 2. Activation energy plot of InGaN quantum well emission. The slope of the straight lines gives an activation energy of 69 meV.

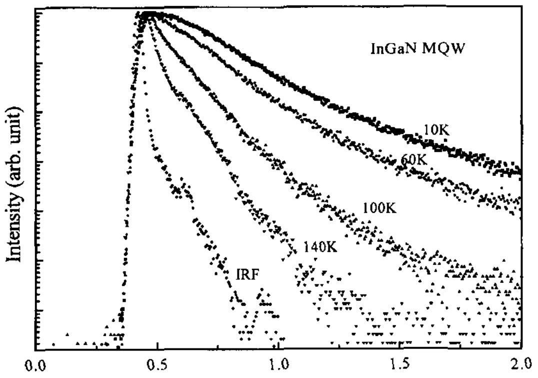

Fig. 3. Time-resolved PL decay profiles of InGaN quantum well emission at various temperatures. The frequency-doubled uv pulses at 290 nm from a synchronously pumped picosecond dye laser were used for photoexcitation. IRF represents the instrument response function of our TCSPC system.

exciton binding energy in GaN type materials is approximately 26 meV. Abnormally large activation energy in the present case indicates that the excitons are not in the free state but they must have been trapped in potential fluctuations in the lattice. Such excitons are generally referred to as localized excitons. The estimated activation energy should correspond to the sum of the exciton binding energy and the potential depth of the trap.

Fig. 3 shows the time-resolved PL decay profiles of InGaN quantum well emission at various temperatures. At 10 K the PL decay time was deduced as 720 ps. For reference we have measured the lifetime of undoped GaN epilayers, which turned out to be about 100 ps at 10 K. The PL lifetimes of GaN and related materials are expected to be very short. The physical processes for the fast radiative relaxation in GaN was suggested to be a strong exciton-acoustic phonon interaction, which gives rise to fast energy relaxation of free excitons to the bottom of the exciton band, leading to generally observed short free-exciton lifetimes. But the PL lifetime in our InGaN MQW is about seven times that of free excitons in undoped GaN. This clearly shows that the excitons in InGaN are not in the free state. The slowdown in the exciton relaxation in InGaN is attributed to the trapping of excitons in potential fluctuations in the lattice. Sugawara [25] has demonstrated theoretically that the lifetimes of such excitons are about 5~7 times those of free excitons. It is remarkable to note t the observed lifetimes of localized excitons in InGaN quantum wells fall exactly in the same range as predicted by Sugawara [25]. We also like to point out that the lifetimes of InGaN epilayers and quantum wells reported by other groups are several times longer than the predictions by theory [25]. Narukawa *et al.* [17] have reported the lifetime of 5 ns for the localized excitons in $In_{0.20}G_{0.80}N/In_{0.05}Ga_{0.95}N$ MQWs. Satake *et al.*[18] have reported the decay time of 1.5 ns for localized excions in 200 nm thick $In_{0.14}Ga_{0.86}N$ epitaxial layers. For a series of $In_{0.22}Ga_{0.78}N/GaN$ MQWs, the lifetimes of ~1.87 ns were observed by Pophristic *et al.* [20] even at room temperature. The relatively short lifetimes of our samples can be rationalized by considering two factors. Firstly it is the structural effect of thin quantum well (10 Å) which results in a strong quantum confinement. Our results show that the electrons and holes in the localized excitons are more strongly bound in thin quantum wells than in the cases of wider quantum wells. Secondly the potential fluctuation due to interface disorder in our samples is much less than the samples prepared by others, indicating that our sample quality is good despite of its thin quantum well structure. As shown in Fig. 3 the lifetime was found to decrease with an increase in temperature. This shows that non-radiative recombination processes come into play as the temperature is increased. This factor should be considered to be important since the actual photonic devices like semiconductor lasers and light emitting diodes are usually operated at hier temperatures. The non-radiative relaxation has deleterious effects on the performance and efficiency of the semiconductor lasers. Recent studies by Narukawa *et al.* [21] have given contradictory and peculiar results that there is no non-radiative transition in their InGaN light emitting diodes. Thus, further analysis of radiative and non-radiative relaxation dynamics and their temperature dependence in InCaN quantum wells will be desirable.

In conclusion, we have studied the steady-state and time-resolved PL of thin quantum wells of $In_{0.13}Ga_{0.87}$ N/GaN MQWs. Their PL dynamics were explained in terms of the radiative recombination of excitons in the localized states. The narrow width of quantum well was found to have a significant effect on the inhomogeneous broadening of the PL linewidth.

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