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Time-resolved photoluminescence studies of InGaN epilayers

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Time-resolved photoluminescence (PL) has been employed to study the optical transitions and their dynamic processes and to evaluate materials quality of InGaN epilayers grown by metalorganic chemical vapor deposition. Our results suggest that the PL emissions in InGaN epilayers result primarily from localized exciton recombination. The localization energies of these localized excitons have been obtained. In relatively lower quality epilayers, the localized exciton recombination lifetime τ, decreases monotonically with an increase of temperature. In high quality epilayers, τ increases with temperature at low temperatures, which is a well-known indication of radiative exciton recombination. Our results demonstrate that time-resolved PL measurements uniquely provide opportunities for the understanding of basic optical processes as well as for identifying high quality materials. © *1996 American Institute of Physics.* [S0003-6951(96)00545-1]

Due to their superior physical properties, the group III-nitride wide bandgap semiconductors have been recognized recently as very important materials for fabricating optoelectronic devices operating in the blue/UV region and electronic devices capable of operating under high-power and high-temperature conditions.[1,2] InGaN has the advantage of tunability of the alloy band gap, allowing greater control over the spectrum of emitted light from visible to near UV. The commercial availability of super-bright blue light emitting diodes (LEDs) and the recent fabrication of blue laser diodes (LDs) based on the InGaN system have demonstrated the tremendous potential of this new ternary alloy system.[3,4] As of yet, many fundamental optical properties of InGaN have not been extensively studies. The physical origins of the emission lines being utilized for light emission in LEDs and LDs are not yet fully understood. Further improvements in the material quality of this ternary alloy system are still necessary. Thus there appears to be a need for a comprehensive study on the properties of the optical transitions and their dynamic processes in InGaN epitaxial layers, which will provide not only a better understanding of fundamental optical properties, but also provide input for new approaches toward the improvement of materials quality as well as the design of particular device structures. In this work, we have employed time-resolved photoluminescence (PL) emission spectroscopy to study the optical transitions in InGaN epilayers with different InN mole fractions. Our results suggest that the PL emissions in these alloys result primarily from localized exciton recombination. Our time-resolved PL data clearly reveal an increase of the localized exciton recombination lifetime with temperature up to 100 K in high quality epilayers, which is an unprecedented observation in III-nitride epilayers, but is a well-known indication of radiative exciton recombination in the well-understood AlGaAs system.

The wurtzite, nominally undoped *n*-type 0.25 μm $In_xGa_{1-x}N$ epilayers were grown on 1.7 μm GaN epilayers using a low pressure metalorganic chemical vapor deposition (MOCVD). The GaN epilayers were deposited on sapphire substrates with thin GaN buffer layers. In this letter, we present results of two representative $In_xGa_{1-x}N$ samples, sample A ($x \approx 0.2$) and sample B ($x \approx 0.12$). The growth procedures of the samples were similar to those described in an earlier work.[5] It was shown there (Ref. 5) that the material quality of sample A was high enough to yield vertical-cavity stimulated emission under a threshold pumping density of about 2.5 MW/cm^2 at room temperature. Sample B was grown very recently and we expect its quality to be superior to sample A due to optimized growth conditions. Low temperature time-resolved PL spectra were measured by using a picosecond laser spectroscopy system with an average output power of about 20 mW, a tunable photon energy up to 4.5 eV, and a spectral resolution of about 0.2 meV. The time resolution of our detection system has been improved compared with our previous setup.[6] A microchannel-plate photomultiplier tube (MCP-PMT) together with a single photon counting system were used to collect time-resolved PL data and the overall time resolution of the detection system is about 20 ps. However, the lifetimes obtained by our new detection system and the previous detection system (with the use of a deconvolution technique) are consistent.

In Fig. 1, we plot the cw PL spectra of samples A and B measured at $T=10$ K. We observe in each sample a single emission band. The peak position in sample $A(B)$is at about 3.07 (3.19) eV with a full linewidth at half-maximum (Δ_{FWHM}) of about 110 (50) meV. The difference in the spectral peak positions in these two samples can be accounted for by the difference in InN fractions. The PL emission linewidth seen in sample A ($\sim$110 meV) here is quite typical and has been observed in the same material grown by different groups.[7,8] However, the linewidth seen in sample B (50 meV at 10 K and 95 meV at 300 K) is among the narrowest ones reported for the InGaN epilayers. The values of Δ_{FWHM} shown in Fig. 1 clearly illustrate that sample B has a

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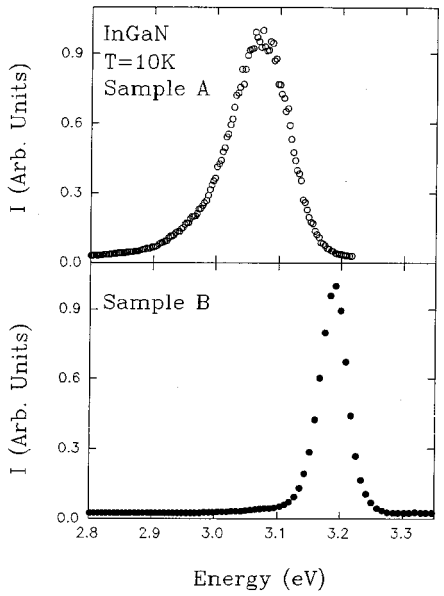

FIG. 1. cw PL emission spectra of $In_xGa_{1-x}N$ epitaxial layers measured at 10 K for sample A ($x \approx 0.2$) and sample B ($x \approx 0.12$).

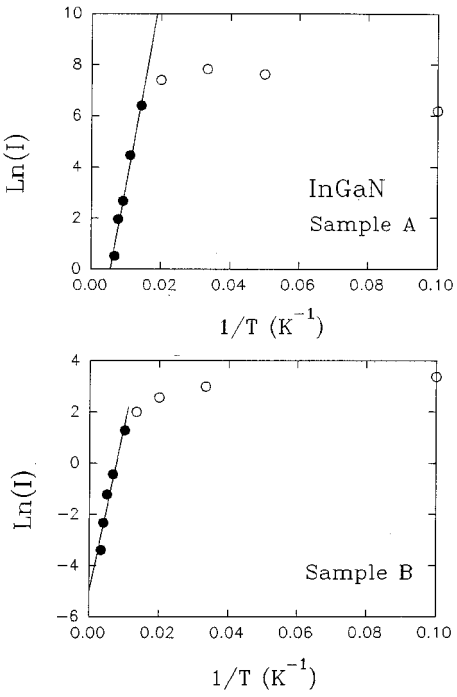

FIG. 2. The Arrhenius plots of the PL emission intensity, ln I vs 1/T for samples A and B.

much higher crystalline quality than sample A. We also find that the PL emission intensities in both samples, I_{emi}, increase superlinearly with excitation intensity, I_{exc}, following a power-law form, $I_{\text{emi}} \propto I_{\text{exc}}^{\beta}$, where the exponent β is about 2.6. The superlinear increase of I_{emi} with I_{exc} indicates that the main emission lines in these InGaN epilayers are of intrinsic nature. The superlinear dependence of the PL emission intensity on the excitation intensity has also been observed previously for the free excitonic transitions in high-quality GaN epilayers grown by MOCVD, which was attributed to enhanced exciton-exciton interactions at high exciton densities.[6]

The PL emission intensity as a function of temperature has also been measured. Figure 2 shows the Arrhenius plots of the PL emission intensities measured in samples A and B, $\ln I$ vs $1/T$. In sample A, the PL intensity increases with temperature below 40 K and then decreases as temperature further increases. In sample B, the PL emission intensity decreases monotonically with temperature. Thermal quenching of the PL emission intensity is due to the formation of fewer excitons at higher temperatures. At $T > 80$ K, the integrated PL intensities in both samples are thermally activated with an activated energy E_0 of about 62 meV for sample A and 56 meV for sample B. It was demonstrated previously that the thermal activation energies of the neutral-donor-bound exciton (I_2) and the free exciton in GaN are very close to their binding energies (~8 and ~20 meV, respectively).[6] Thus the observed activation energies E_0 preclude the assignment of the dominant emission lines in these samples to a bound exciton or a free exciton recombination, but rather support a previous assignment of a localized exciton recombination.[8] In a semiconductor alloy, the exciton localization is caused by energy fluctuations in the band edge induced by alloy disorder. The measured activation energies represent the localization energies of the localized excitons in the InGaN samples. In such a context, the localized exciton emission linewidth is also induced by the energy fluctuations in the band edge, which should then be of the same order as the excition localization energy. This is in fact the case for sample B, in which the localized exciton emission linewidth is 50 meV and the exciton localization energy is 56 meV. However, in sample A, the PL emission linewidth (~110 meV) is about a factor of 2 larger than the exciton localization energy (~62 meV). We believe that dislocations caused by lattice mismatch between InN and GaN and native defects also contributed to the linewidth broadening in sample A. This is supported by the results shown in Fig. 2(a), which shows that the PL intensity actually increases with an increase of temperature at $T < 40$ K, and hence points to the existence of impurity related optical transitions located probably in the lower energy shoulder of the localized exciton emission line. The impurity related optical channels can pump the localized exciton transition at moderate temperatures. The monotonic decrease of the PL intensity with temperature shown in Fig. 2(b) indicates that contributions from other impurity related optical transitions are negligible in sample B and hence points to the higher purity as well as higher crystalline quality of sample B. These results are consistent with our recent efforts in optimizing the growth conditions for $In_xGa_{1-x}N$ epilayers including sample B.

The recombination dynamics of the PL emissions in these epilayers have been studied. Figure 3 shows the temporal responses of the localized exciton recombination resulting from sample B measured at the spectral peak positions at three different temperatures $T = 10$, 100, and 200 K. The detection system response to the laser pulses is also indicated as ''system,'' which is about 20 ps. As shown in Fig. 3, the PL decay in $In_xGa_{1-x}N$ epilayers can be described quite well by a single exponential at all temperatures. The PL recombination lifetimes, τ, were measured at the spectral peak positions from 10 K to room temperature,

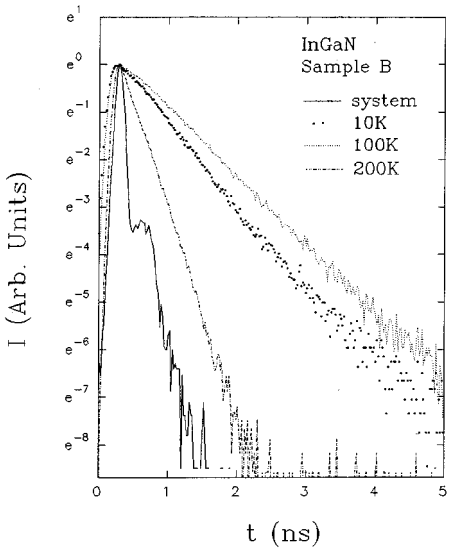

FIG. 3. The temporal responses of the PL measured at the spectral peak positions in sample B at three representative temperatures. The system response to the laser pulses is indicated as ''system'' and is about 20 ps.

which are shown in Fig. 4 for both samples. At room temperature, the values of τ in both samples are about 0.1 ns. However, the temperature dependencies of τ for both samples are quite different. In sample A, the recombination lifetime is about 0.33 ns at low temperatures ($T<30$ K) and decreases gradually with temperature, which is most likely due to the increased nonradiative recombination rate at higher temperatures. In a remarkable contrast, in sample B, the localized exciton lifetime is about 0.53 ns at $T<40$ K and the *increases* almost linearly with temperature up to 100 K. To our knowledge, this is the first time that such a linear increase of the localized exciton recombination lifetime with temperature has been recorded for the InGaN alloy system. A linear behavior of τ vs T at low temperatures has been observed previously for exciton recombination in GaAs/Al_xGa_{1-x}As multiple quantum wells (MQW) and is

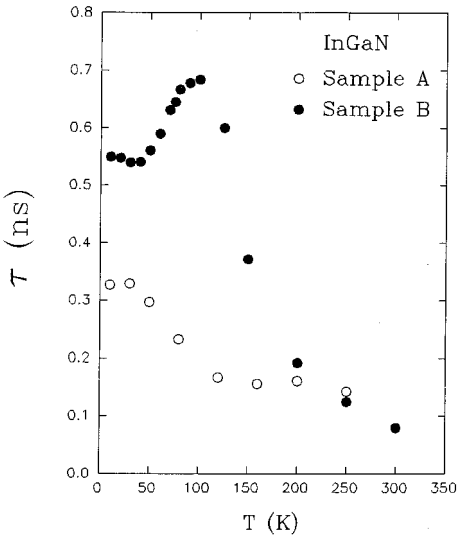

FIG. 4. The temperature dependencies of the PL recombination lifetimes measured at the spectral peak positions of samples A and B.

now regarded as a hallmark of radiative recombination in MQW.[9] Thus our time-resolved PL data show that the radiative recombination is a dominant process in sample B at low temperatures, which again demonstrates the high quality of sample B used here. The longest radiative lifetime seen in sample B is 0.70 ns at 100 K, which is more than two times larger than a value of 0.33 ns seen in sample A and is the longest lifetime reported so far for intrinsic transitions in InGaN alloys.

The low temperature PL recombination lifetimes in both samples are also functions of emission energy and excitation intensity (not shown). In sample A, at 10 K, τ is 0.38 ns at 3.01 eV and decreases with emission energy to about 0.1 ns at 3.15 eV. In sample B, at 10 K, τ is about 0.75 ns at 3.15 eV and decreases with emission energy to about 0.51 ns at 3.21 eV. Such an emission energy dependence of τ is a characteristic of localized excitons in semiconductor alloys.[10] On the other hand, τ in both samples decrease with an increase of excitation intensity, similar to the behavior of the free excitons in GaN.[6] Since the PL quantum yield increases with a decrease of the radiative recombination lifetime, the excitation intensity dependence of τ can also account for the superlinear increase of PL emission intensity with excitation intensity observed in these samples.

In summary, we have employed time-resolved PL to study optical transitions and to evaluate the quality of InGaN epilayers grown by MOCVD. Our results suggest that the PL emissions in InGaN epilayers result primarily from localized exciton recombination. The localization energies of these localized excitons are around 60 meV, which corresponds to a composition fluctuation in In of about ± 0.03. In epilayers of relatively lower quality, the localized exciton recombination lifetime decreases monotonically with an increase of temperature. In high quality epilayers, the localized exciton recombination lifetime increases almost linearly with temperature in certain low temperature regions. A recombination lifetime of 0.7 ns and a narrowest linewidth of 50 meV at 10 K (95 meV at 300 K) have been observed in our high quality epilayers, which are among the longest lifetimes and narrowest linewidths reported for the InGaN system.

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