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By using a “nanoscale aperture” method, we studied the dynamics of localized excitons in active InGaN layers in an InGaN/GaN multiple quantum well (MQW). Local photoluminescence (PL) from local excitation shows very different characteristics from far-field luminescence. PL spectra from these nanoapertures are, in general, blueshifted relative to the spectra acquired in the unmasked regions. The spectra vary from aperture to aperture, reflecting the spatial fluctuation of the In composition in the MQW. In addition, one typically observes spectra with clearly resolved multiple peaks from the small apertures, which may be attributed to compositional and size fluctuations over a length scale longer than the exciton diameter but smaller than the aperture size. The excitation intensity dependence of PL indicates a band-filling effect in the multiple-peak structure. Finally, time-resolved studies reveal a very interesting spectral weight shifting between the higher-energy peaks and the lower-energy peaks. © 1998 American Vacuum Society. [S0734-211X(98)08104-9]

The recent breakthrough in the development of nitride-based blue and green light-emitting diodes (LEDs) and laser diodes (LDs) opens a new era in modern optoelectronic technology.¹⁻³ However, many important materials issues remain to be investigated. The lack of detailed understanding of the optoelectronic processes in these GaN-based devices presents scientific challenges and research opportunities. An important issue is related to the InGaN active layers in InGaN/GaN quantum wells.¹ Due to the large mismatch of the lattice constant between GaN and InN, it is very difficult to grow homogeneous InGaN layers. Phase separation into regions of different In composition is the rule rather than exception.^{4,5} In such highly inhomogeneous active layers, optoelectronic processes such as carrier transport and exciton dynamics are highly complex. It has been suggested that the localized In-rich regions act as quantum dots (QDs), which are responsible for the lasing action in GaN-based LDs.⁶⁻⁸

The samples used in this study are InGaN multiple quantum wells (MQWs) grown on a 1.5- μm -thick GaN:Si layer deposited on a sapphire (0001) substrate by low-pressure metalorganic chemical vapor deposition. The MQW structure consists of five periods of 3.5-nm-thick $\text{In}_{0.13}\text{Ga}_{0.87}\text{N}$ Si-doped quantum wells and 7-nm-thick $\text{In}_{0.03}\text{Ga}_{0.97}\text{N}$ Si-doped barrier layers.^{9,10} A 16-nm GaN:ud (undoped GaN) cap is grown on top of the MQW structures. The dynamic behavior of localized excitons in the InGaN MQW is studied using a “nanoscale aperture” method. Such a method has been used previously by others to study optical properties of single GaAs QDs.^{11,12} However, instead of using electron-beam lithography, here we use a much simpler way to create nanoapertures by utilizing polystyrene (PS) beads.

The fabrication of the nanoscale apertures is shown schematically in Fig. 1. A suspension 0.3- or 0.6- μm -diam poly-

styrene beads diluted with alcohol was dropped onto the sample. After the alcohol evaporated, sparsely distributed PS beads were then left on the sample surface. After the deposition of 100 nm Al thin films followed by the removal of the PS beads using dichloromethane solution and ultrasonic agitation, an Al metal film with small apertures is formed. Most of the apertures have the same size as the diameter of the PS bead as revealed by the atomic force microscope (AFM) image (Fig. 2). However, some apertures are larger with an apparent shape due to the aggregate of a few PS beads. Using this method, we create apertures with size ranging from 0.3 to 3 μm . The holes were spaced sufficiently far apart to allow optical probing of a single hole. The photoluminescence (PL) was excited by temperatures from 10 to 300 K through a given hole with a picosecond laser system, and the PL was detected from the same hole with 1.33-m-long spectrometer and microchannel-plate photomultiplier tube together with a single-photon counting system.⁵ Hence, the excitation and PL detection was performed via the same aperture.

The cw PL spectra acquired at 10 K from the submicron apertures and the spectrum from the unmasked region are shown in Fig. 3. It can be clearly seen that the PL peak positions of the masked region shift to higher energy as compared to the unmasked region. This blueshift trend can be explained in the context of exciton diffusion out of the aperture region, from higher-energy sites to lower-energy sites. Excitons localized in random wells induced by composition fluctuation will transfer from higher-energy site to lower-energy site by thermal relaxation. Those excitons which diffuse out of the aperture then will have lower energies and will not be detected due to the opaque Al cover. While for the unmasked sample, PL is dominant from the lowest-energy region due to the localization effect. Thus, PL emitted from an aperture will have a relatively higher-energy portion,

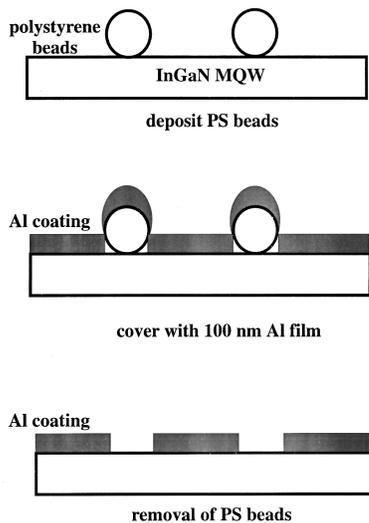


FIG. 1. Schematic diagrams of the preparation of the nanoscale apertures.

i.e., exhibit a blueshift. In the masked sample, if there is no such potential difference, or no composition fluctuation when excitons diffuse out of the aperture, there will be no peak position shift of the spectra. Therefore, blueshift of the spectra indicates there is a composition fluctuation in the MQW and excitons are localized to the lower-band-gap regions of MQW corresponding to higher In composition. It also indicated that the exciton diffusion and localization process covers at least $1 \mu\text{m}$.

One also sees in Fig. 3 multiple peaks on the spectra from the masked area. The appearance of several PL peaks in the spectra is not due to an interference effect, because there is no spacing in the structure that will give rise to the peak energy separation in the wavelength region we are studying. These peaks are also not phonon replicas, as has been observed in other studies on InGaN/GaN and GaN/AlGaIn MQWs. The longitudinal optical phonon energies of InN and GaN are 86 and 91 meV, respectively,¹³ while the energy separations between neighboring peaks in our spectra are, typically, about 30 meV, though the separations vary from one to another. The multiple-peak structure may be attributed to the compositional and size fluctuation over a length scale longer than the exciton diameter but smaller than the aper-

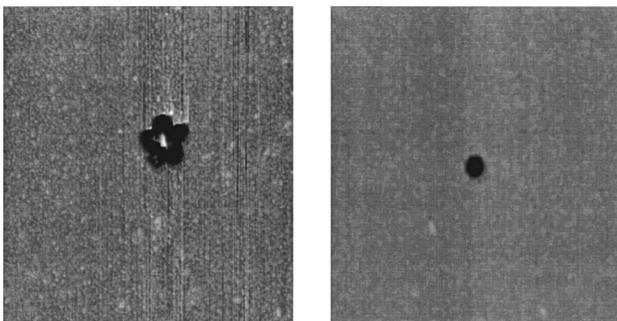


FIG. 2. $2.5 \mu\text{m} \times 5 \mu\text{m}$ AFM images showing apertures fabricated using PS beads of $0.3 \mu\text{m}$ size. Most of the apertures made are $0.3 \mu\text{m}$, in some cases, apertures are of $1 \mu\text{m}$ size due to clustering of the PS beads.

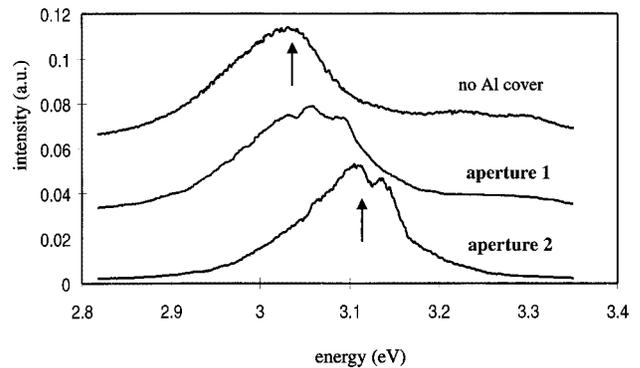


FIG. 3. Photoluminescence spectra taken from unmasked and masked regions.

ture size. It is interesting to note that in contrast to the Gammon and co-workers work, where sharp peaks from single GaAs quantum dots with a linewidth as small as $80 \mu\text{eV}$ can be resolved,^{11,12} here the smallest linewidth is about 20 meV. The reasons for this may be due to the following. First, the size of the aperture here is not as small as in the Gammon and co-workers work. Second, the Bohr radius of the excitons in GaN is about 3.4 nm,⁴ much smaller than that in GaAs, i.e., 20 nm.¹¹ Third, in their work, the excitons are formed due to the well width fluctuations of the quantum wells in GaAs, which are of a lateral size larger than 20 nm.¹¹ In InGaN MQW the composition fluctuations are expected to be of the size of several nanometers, consequently, the exciton localization effect will be much stronger than that in GaAs quantum dots, and give rise to broader peaks due in part to localization and in part due to phonon coupling to higher-energy phonons in InGaN.

Figure 4 shows PL spectra from the same aperture at 10 K for two different excitation intensities. The excitation intensity I_{exc} is proportional to 10^{-D} . The arrows indicate the multiple peak positions of the PL spectra. It is clear that the multiple emission peak positions remain at the same energies when changing the intensity, while the spectral weight shifts toward higher energies with increasing intensity. This excitation intensity dependence indicates a band-edge-filling effect of the multiple-peak structure, where the band edge is due to the local fluctuation of In composition.

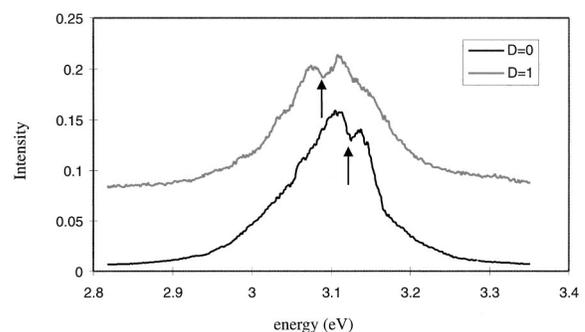


FIG. 4. Photoluminescence spectra from the same aperture with different excitation intensities.

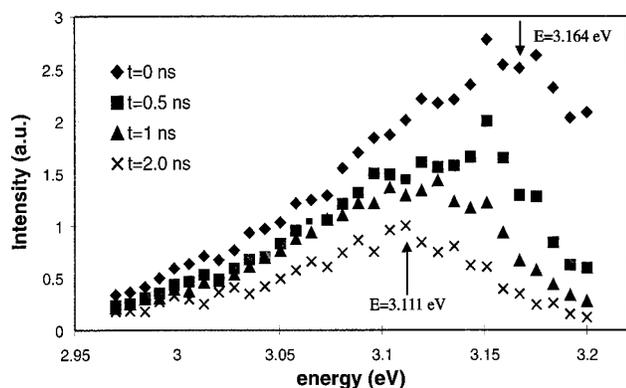


Fig. 5. Time-resolved photoluminescence spectra from the same aperture as in Fig. 4 showing the spectral shifting from high energy to lower energy with increasing delay time.

Time-resolved PL spectra have also been measured on both masked and unmasked regions to study the dynamics of the localized excitons. Figure 5 shows time-resolved emission spectra of the 3.164 eV emission line measured on the same aperture as in Fig. 4 at 10 K at several representative delay times. The arrows in Fig. 5 indicate the spectral peak positions at different delay times. It can be seen that the spectral peak position shifts toward lower energies as the delay time increases and the linewidth of the emission line also increases with delay times. This luminescence spectral shift with delay time is a direct consequence of a strong emission energy dependence of the recombination lifetime. If the optical transition involves only a single exciton energy level, the spectral shift with delay time and the variation of the recombination lifetime with emission energy are not expected. Thus, these results indicate that localized exciton energy levels in the aperture under study have a distribution. With the increasing delay time, the localized excitons are transferred from high-energy levels to low-lying levels within the aperture region.

Two important effects caused by this alloy fluctuation and local excitation/detection of microapertures have to be considered. First, in energy space, excitons relax towards the lower-energy regions due to the exciton localization effect. Second, in real space, excitons simultaneously diffuse away from the aperture due to the presence of the microapertures. Thus, only those excitons which radiatively recombine within the apertures will be detected in this special local excitation/detection configuration. This is the key difference between our results obtained here and previous results of unmasked samples. For samples without apertures, the cw

PL emission spectra are always dominated from those excitons localized in the lower-energy regions, reflecting the fact that exciton localization and diffusion are very fast processes.

In conclusion, a “microaperture” method has been employed to study the dynamics of the localized excitons in InGa/GaN MQWs. The cw spectra from masked and unmasked regions revealed that excitons are localized into spatially different regions due to the phase separation into different In composition regions in InGaN and the resulting composition fluctuation in quantum wells. The blueshift trend and the spectral shift with delay time have demonstrated that the localized excitons are transferred spatially from high potential energy sites to low potential sites and temporally from high-energy levels to low-energy levels.

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