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Band gap renormalization and carrier localization effects in InGaN/GaN quantum-wells light emitting diodes with Si doped barriers

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The optical properties of two kinds of InGaN/GaN quantum-wells light emitting diodes, one of which was doped with Si in barriers while the other was not, are comparatively investigated using time-integrated photoluminescence and time-resolved photoluminescence techniques. The results clearly demonstrate the coexistence of the band gap renormalization and phase-space filling effect in the structures with Si doped barriers. It is surprisingly found that photogenerated carriers in the intentionally undoped structures decay nonexponentially, whereas carriers in the Si doped ones exhibit a well exponential time evolution. A new model developed by O. Rubel, S. D. Baranovskii, K. Hantke, J. D. Heber, J. Koch, P. Thomas, J. M. Marshall, W. Stolz, and W. H. Rühle [J. Optoelectron. Adv. Mater. **7**, 115 (2005)] was used to simulate the decay curves of the photogenerated carriers in both structures, which enables us to determine the localization length of the photogenerated carriers in the structures. It is found that the Si doping in the barriers not only leads to remarkable many-body effects but also significantly affects the carrier recombination dynamics in InGaN/GaN layered heterostructures. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2168035]

Strained InGaN-based quantum wells (QWs) are currently of great interest due to their applications in the short wavelength light emitting diodes (LEDs) and laser diodes as light emitting active layer.[1] One idiosyncratic feature in present InGaN QWs is the carrier localization due to the apparent local compositional fluctuations in the In and Ga compositions and the strain-induced piezoelectric field.[2] Several groups have reported that Si doping in the GaN barrier layers may result in a decrease of the carrier localization to a certain extent at the potential fluctuations in the InGaN active layers[3] and leads to a partial screening of the strain-induced piezoelectric field.[4] However, to our knowledge, band gap renormalization (BGR) and phase space filling (PSF) effects have not been observed in InGaN QWs with Si doped barrier layers. Furthermore, the influence of Si doping on the recombination dynamics of carriers in InGaN QWs is still unclear. Therefore, a detailed investigation of the BGR and PSF effects is highly desirable due to both requirements for performance improvement of InGaN-QWs based light emitting devices and understanding of the associated fundamental physics.

In this letter, we present the results obtained from a comparative study on the optical properties of two kinds of InGaN/GaN quantum wells LEDs with identical geometric structure and composition, one of which was doped with Si in the barriers while the other was not. The experiment results indicate that Si doping can give rise to a significant reduction of the fundamental gap energy which is the so-called BGR effect. In addition, the PSF effect was also observed in the doped structures. It is also found that photogenerated carriers in the doped sample possess a shorter localization length. Two InGaN/GaN-QWs LED structures having identical structure and the same indium concentration, labeled by A and B, were grown on *c*-plane sapphire substrate by metalorganic chemical vapor deposition. Each sample consists of a 3 μm n^+-GaN layer, an active layer of five periods InGaN/GaN QWs and a 200 nm p^+-GaN layer. Thickness of the well (barrier) layer is 3 nm (12 nm). The only difference between these two samples is that the barrier layers of QWs in sample A were intentionally undoped while the barrier layers of sample B were doped with Si to about 10^{18} cm^{-3}. For the time-integrated photoluminescence (PL) measurements, the 325 nm UV line from a Kimmon He–Cd continuous wave laser with a maximum output power of 34 mW was used to excite the samples which were mounted on the cold finger of a Janis SHI closed circle refrigerator with a varying temperature range from 5 K to room temperature. The luminescence signal is collected by lens and dispersed by a Spex 750 M spectrometer and finally detected by a Hamamatsu R928 photomultiplier tube interfaced with lock-in amplifier. For the time-resolved photoluminescence measurements, we employed the frequency doubled femtosecond pulses (λ_{exc}=355 nm) from a Ti:sapphire laser with repetition rate of 82 MHz to excite the samples which were loaded into a 77 K Dewar. The luminescence decay was detected with a Hamamatsu streak camera with a temporal resolution of ~20 ps.

Figure 1 shows the semilogarithmic PL spectra of the two samples at 5 K under identical experimental conditions (i.e., same excitation power of 33 mW). Several spectral features can be identified. First, the peak intensities of the PL spectra from both samples are almost equal. Furthermore, the PL spectra of both samples have very similar structures at

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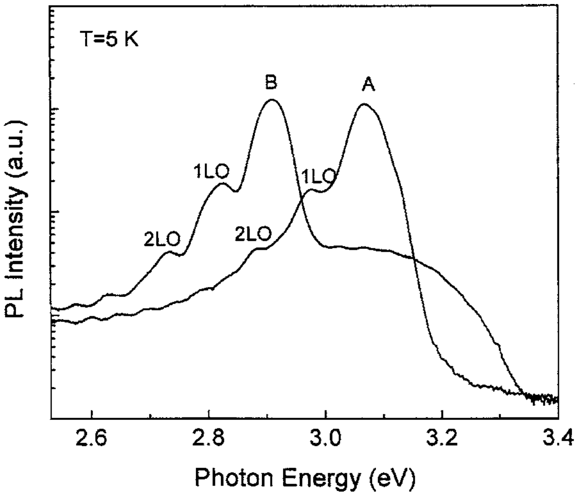

FIG. 1. Measured 5 K PL spectra of samples A and B under excitation of UV laser with power of 33 mW.

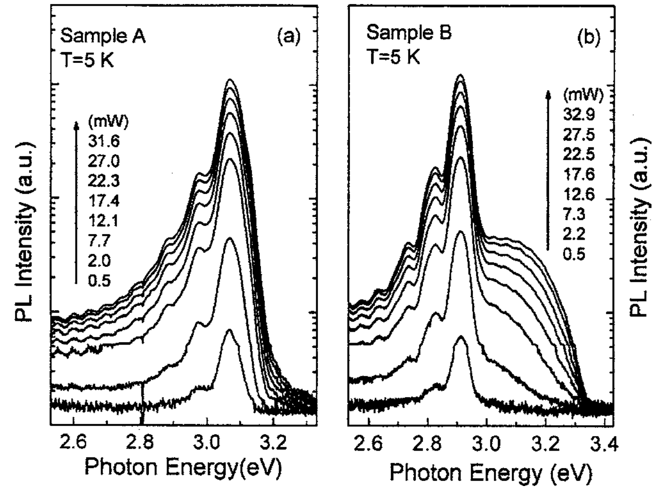

FIG. 2. Excitation power dependent PL spectra at 5 K (a) for sample A and (b) for sample B.

lower energy side of their principal peak. There are at least two LO phonon sidebands (referred to as 1LO and 2LO) which can be resolved. The energy separations between the principal peaks and 1LO as well as between 1LO and 2LO are identical and can be determined to be 90.5±0.5 meV which is very closed to the characteristic LO phonon energy of GaN (~91 meV).[5] Second, the principal peak (located at ~2.910 eV) and its LO-phonon sidebands of sample B exhibit a redshift of about 158 meV with respect to the corresponding peaks of sample A. Such a redshift should be characteristic of the BGR effect due to the screening of free carriers.[6] The BGR effect has been observed in Si doped GaN and AlN epilayers.[7,8] In the InGaN/GaN QWs sample with Si doped barriers studied in the present work, a large mount of electrons from the Si donors in the barrier layers can form a high density of Fermi gas in the wells. Exchange and correlation of electrons results in renormalization of the band gap which is manifested by a redshift of the luminescent band gap. An empirical expression, $\Delta E_{\mathrm{PL}}=-Kn^{1/3}$, where K is a proportionality constant depending on materials and n is the carrier concentration, is usually used to evaluate the BGR shift. If ΔE_{PL} is simply assumed to be proportional to Si doping concentration, K of InGaN/GaN QWs is estimated to be 1.58×10^{-4} meV cm, which is higher by several folds than that of doped GaN epilayers.[7,9] Larger BGR effect is attributed to enhancement of carrier localization degree in the well layers due to Si doping in the barrier layers, as proved later.

Another spectral feature in Fig. 1 is that a broad shoulder appears at the higher energy side of the PL spectra of sample B. This shoulder is attributed to the filling of the higher energy states by electrons releasing from the Si donors in the sample. It is well known that as the carrier concentration increases, electronic states at higher energies will be occupied according to the Pauli Exclusion Principle, and thus the Fermi level goes up. That is the so-called phase space filling effect. A possible spectral feature of the PSF effect is that the high energy edge of the PL peak expands towards high energy direction or new structures appear at the higher energy side of the PL peak.[10] In order to verify that the PSF effect is really observed in the Si doped sample, excitation power-dependent PL measurements were performed. The results are shown in Fig. 2. Indeed, a broad shoulder gradually appears at the higher energy side of the PL spectra of sample B with increasing the excitation power. For the intentionally undoped sample (sample A), only small blueshift of the high energy edge of the PL spectra is observed. The broad shoulder at the higher energy side of the PL spectra of sample B is thus attributed to the radiative recombination of electrons occupying higher energy states and the optically generated holes. The involved electrons are mainly from the Si donors. We also note that the PL peak energies of both samples remain nearly unchanged and even the excitation power is increased from 0.5 mW to higher than 30 mW. This means that the piezoelectric field frequently observed in strained InGaN/GaN QWs is successfully suppressed in the samples studied in the present work.

To get more information, the time evolution of PL intensity of both samples at 77 K was measured, as indicated in Fig. 3. The intensity of the PL signal from sample A exhibits a nonexponential decay behavior, which is a typical characteristic of a disorder system. In contrast to the sample A case, the time evolution of the PL signal from sample B puzzlingly obeys the single exponential decay of the exciton picture.

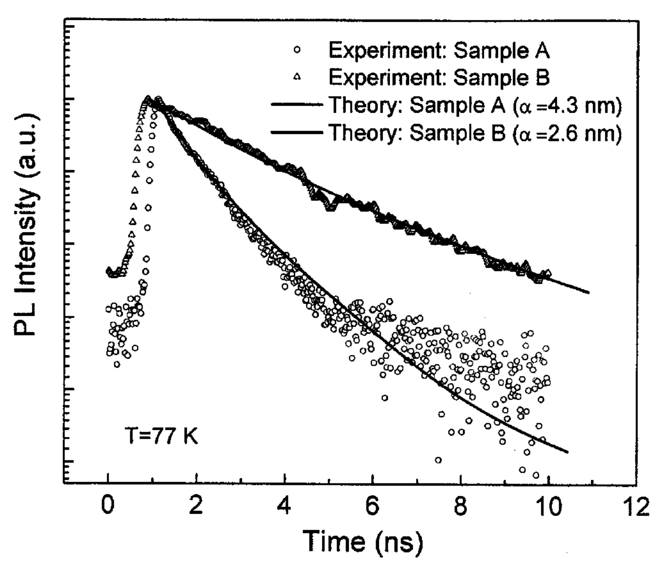

FIG. 3. Measured PL decay profiles (empty symbols) at 77 K for samples A and B. The solid lines are the fitting curves using the model newly developed by Rubel *et al.* (see Ref. 11).

However, it is hard to believe the existence of excitons in sample B whose GaN barriers were heavily doped with Si atoms. For interpretation of this puzzling observation, we employ a new theoretical model developed by Rubel *et al.*[11] to simulate the experimental curves. The theory is based on the set of rate equations, in which electrons and holes involved in luminescence process are treated as uncorrelated carriers rather than in the form of excitons. According to this new model, for a semiconductor system with strong enough disorder, charge carriers can be instantaneously captured into localized states of the system after their photogeneration. From these states the carriers can either radiatively recombine or perform a phonon-assisted hopping transition to other localized states. Since both these processes take place in the localized states, their probabilities depend exponentially on the distances involved.[11] Behaviors of carriers' recombination dynamics are thus essentially determined by carriers' localization lengths.

Using this model, a fitting to the experimental curves shown in Fig. 3 was carried out. The solid curves in Fig. 3 are the fitting results when the values of parameters $\tau_0 = 1$ ns, $\nu_0 = 10^{10}$ s^{-1}, $kT = 6.64$ meV, $\varepsilon_0 = 8$ meV were adopted. The definitions of these parameters can be found in Ref. 11. Obtained carrier localization lengths are 4.3 nm for sample A and 2.6 nm for sample B, respectively. Clearly, sample B (Si doped) has a shorter localization length. Since the localization length is a most important parameter characterizing the disorder potential, the results show that Si doping in the barrier layers of InGaN/GaN QWs can lead to deeper localization of carriers and thus reduce hopping rate of carriers between different localized states. The deeper localization of carriers at potential minima certainly results in a redshift of luminescence peak of the system. That is why a larger redshift in the PL peak than expected is observed by the BGR effect. As mentioned earlier, for a system with strong enough disorder, recombination times of photogenerated carriers have a broad distribution since they depend exponentially on the distances R between recombination partners. It reads[11]

$$\tau_r(R) = \tau_0 \exp(2R/\alpha), \tag{1}$$

where τ_0 is a time constant which depends on the particular recombination mechanism and is of the order of excitonic radiative lifetime, and α is the localization length. According to Eq. (1), the recombination time is exponentially dependent on the inverse of the localization length. Observed longer lifetime for the Si doped sample, as shown in Fig. 3, is consistent with the theoretical prediction. This result also supports the conclusion that the Si doping in the barrier layers is able to enhance the localization degree of carriers in the potential minima of InGaN well layers.

In summary, the influence of Si doping in the barriers of InGaN/GaN QWs LEDs on optical properties of the structures is investigated. Noticeable redshift of the band-edge emission peak is observed for the Si doped samples and is mainly attributed to the band gap renormalization due to the interaction of electrons in the wells. The excitation-intensity dependence experiment shows evidence of the phase space filling effect in the doped samples. The radiative dynamics of the photogenerated carriers in the samples is also studied. Measured decay curves of the PL intensities were simulated by means of a newly developed theoretical model to determine the localization length of carriers. It is found that for the Si doped sample, the photogenerated carriers possess a shorter localization length. This means that Si doping in the barrier layers of InGaN/GaN QWs is helpful to enhance luminescence efficiency of the structures.

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