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We employ photoluminescence (PL) and time-resolved PL to study exciton localization effect in InGaN epilayers. By measuring the exciton decay time as a function of the monitored emission energy at different temperatures, we have found unusual behaviour of the energy dependence in the PL decay process. At low temperature, the measured PL decay time increases with the emission energy. It decreases with the emission energy at 200 K, and remains nearly constant at the intermediate temperature of 120 K. We have studied the dot size effect on the radiative recombination time by calculating the temperature dependence of the exciton recombination lifetime in quantum dots, and have found that the observed behaviour can be well correlated to the exciton localization in quantum dots. This suggestion is further supported by steady state PL results.

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It has been widely demonstrated that the InGaN-based light-emitting devices are highly efficient with very low thresholds. The high efficiency of the light emission is believed to be closely related with the effect of carrier localization.[1,2] The main radiative recombination has been attributed to the emission of excitons localized in the In-rich regions of the InGaN active layers caused by a phase separation. The In-rich regions act as self-assembled quantum dots (QDs). Therefore, to provide experimental evidences for the QD formation in the InGaN layer has been a continual effort in the literature.

The first direct observation of QDs formation in the InGaN layer was reported by Narukawa *et al.*[3] The authors studied transmission electron microscopy (TEM) and energy dispersive x-ray (EDX) microanalysis on cross sections of the InGaN multi-quantum-well (MQWs) and revealed a variation in the In concentration in the well layers. The TEM images showed rows of dark spots in the MQWs, which were assigned to quantum dots. From high-resolution electron microscopy, Nistor *et al.*[4] also provided direct evidence of the QDs formation in a single 280-nm-thick layer of $In_{0.22}Ga_{0.78}N$.

Optically, O'Donnell *et al.*[5] studied the scalabilities of the Stokes shift in the measurements of absorption and emission spectroscopy, and reported the first direct observation of phase separation in an InGaN quantum well by optical spectroscopy. The authors concluded that the phase separation leads to the formation of a nanostructure of nearly pure InN quantum dots. Whereas in the study of Lemos *et al.*[6] it was reported that the In-rich separated phases with nearly the same indium composition of 0.8 in all their samples. The authors demonstrated that the emission of InGaN alloy is due to the quantum confinement effects in the phase-separated In-rich quantum dots. Similar results were also reported in AlInGaN alloys.[7] More recently, Li *et al.*[8] measured the temperature-dependent photoluminescence (PL) of InGaN ternary alloy. The observed anomalous temperature dependence of the PL peak energy was attributed to the thermal redistribution of localized excitons among different potential minima.

In this Letter, we employ PL and time-resolved photoluminescence (TRPL) to study the exciton localization effect in InGaN epilayers. The observed abnormal energy dependence of photoluminescence decay time can be well attributed to the exciton localization in quantum dots. The numerical calculation of the dot-size effect on the radiative recombination time is in agreement *qualitatively* with our experimental results.

The InGaN epilayers used here were grown on sapphire (0001) substrates by metal–organic chemical-vapour deposition (MOCVD). A low-temperature GaN buffer was deposited first, and followed by deposition of 0.5-μm GaN grown at 1030°C. InGaN epilayers with different thicknesses were then grown on the GaN layer under a low pressure of 76 Torr and the growth temperature was 820°C. Trimethylgllium (TMG), ethyldimethylindium (EDMIn) and ammonia were used as precursors, and SiH_4 was used for n-type doping. The indium composition of the samples

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was determined by an energy dispersive system (EDS) and x-ray diffraction (XRD) and estimated to be 0.22. The high indium composition favours the phase separation, as illustrated by high-resolution TEM images (not shown). For the TRPL measurements, the PL was excited by frequency-doubled ($\lambda = 360\,\mathrm{nm}$) laser pulses from a Ti:sapphire mode-locked femtosecond laser and the time-correlated signals were analysed by a two-dimensional (2D) synchronous streak camera with an overall resolution of less than 15 ps. The cw PL measurement was performed using a combination of a cooled GaAs PMT and a Data-Link electronic system.

Figure 1(a) shows the measured PL decay time as a function of the monitored emission energies at three typical temperatures for one of our InGaN samples (with thickness of $0.4\,\mu\mathrm{m}$). At a low temperature of 12 K, the measured PL decay time increases with the emission energy. However, the measured PL decay time decreases with the emission energy when temperature is increased to 200 K. At an intermediate temperature of 120 K, the decay time remains nearly constant. To eliminate the non-radiative recombination effect from the measured PL decay dynamics, we have studied the radiative and non-radiative recombination dynamics by combining the PL and TRPL results.[9] The values of the radiative lifetime τ_r and non-radiative lifetime τ_{nr} can be deduced from the measured PL decay (τ_{PL}) and PL intensity as a function of temperature using the relation $\eta_{\mathrm{int}} = 1/(1 + \tau_r/\tau_{nr})$, where η_{int} is an internal quantum efficiency and set to be unity at 12 K, as the radiative recombination is dominated at low temperature. In Fig. 1(b) the deduced radiative lifetimes are plotted as a function of the emission energies at three temperatures. The results are very similar to the experimental observation shown in Fig. 1(a).

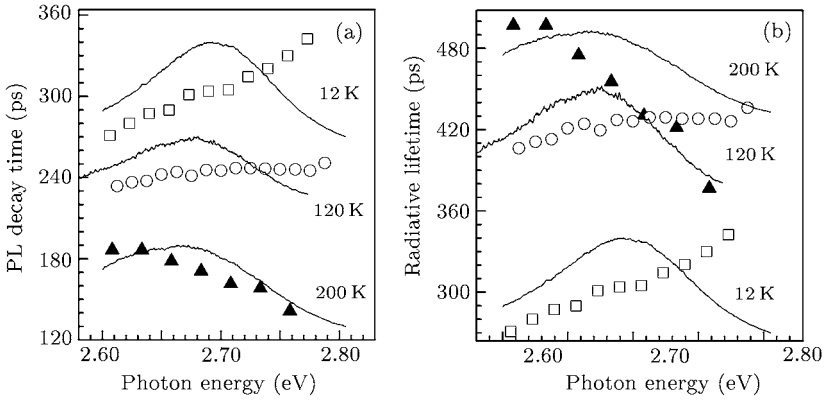

Fig. 1. (a) Measured PL decay time as a function of the monitored emission energies at three typical temperatures for the InGaN epilayer of $0.4\,\mu\mathrm{m}$ thick. (b) Deduced exciton radiative lifetime as a function of the emission energies. The solid curves are the time-integrated PL spectra.

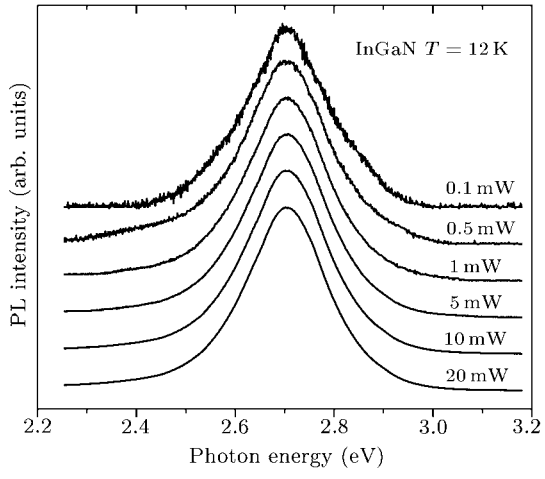

Fig. 2. PL spectra of the InGaN sample at 12 K under different excitation powers. All the spectra are normalized and shifted vertically for clarity.

The emission energy dependence of the PL decay time at low temperature has been widely used to characterize the exciton localization. For example, in the case of exciton localized in the band tail states,[10] the decay time increases with the decrease of the emission energy. This is because the decay of localized exciton is not only due to the radiative recombination but also due to the transfer process to the band tail states. This phenomenon was reported previously in the semiconductor nitride alloys, such as InGaN and GaNAs.[11,12] The localization was attributed to the random distribution of alloy compositions and/or quantum well-width fluctuations. Obviously, our results differ significantly from the above exciton localization effect. We believe that the observed unusual behaviour is correlated with the random distribution of the potential minima due to the quantum dots formation in our InGaN sample. Figure 2 shows the PL spectra of the InGaN sample at 12 K under different excitation powers. It can be seen that the line-shape of the PL is Gaussian-like, in agreement with the Gaussian-like size distribution in the

self-assembled QD structure. Moreover, the PL peak position does not change in the whole excitation range from 0.01 mW to 20 mW. This is in contrast to the case of exciton localization in the band-tail states, where the blue-shift of the PL peak with excitation intensity is often observed.[13,14]

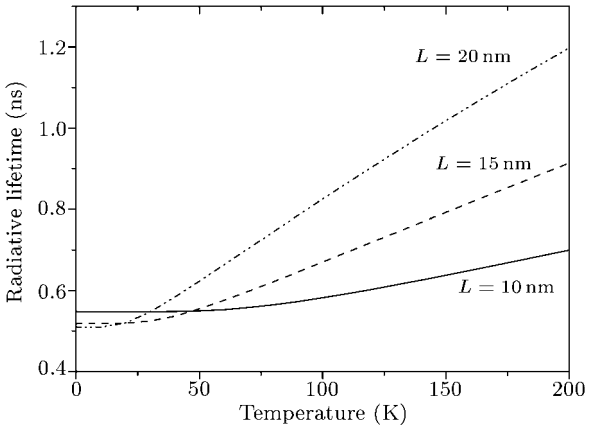

Fig. 3. Calculated temperature dependence of the exciton recombination lifetime in quantum dots for the dot sizes of 10, 15, and 20 nm, respectively.

Now we further discuss the dot-size effect on the radiative recombination time by calculating the temperature dependence of the exciton recombination lifetime in a quantum-dot structure. The calculation is within the framework of effective mass approximation using the following Schrödinger equation, similar to the work done by Goton *et al.*:[15]

$$\left(-\frac{\hbar^2}{2m_e}\nabla_e^2-\frac{\hbar^2}{2m_h}\nabla_h^2+\Delta V_e(\boldsymbol{r}_e)+\Delta V_h(\boldsymbol{r}_h)\right)\Psi = E\Psi, \tag{1}$$

where $\Delta V_e(\boldsymbol{r}_e)$ and $\Delta V_h(\boldsymbol{r}_h)$ are spatial distributions of the potential for electrons and holes, respectively. In the calculation, formation of InN QDs in InGaN matrix is assumed to exist.[16] The thermal population of each exciton state as well as its oscillator strength are considered. In addition, we neglect the inter-dot carrier transfer process and electron-hole Coulomb interaction for the sake of simplicity. The results of the calculation are shown in Fig. 3 for the dot sizes of 10, 15 and 20 nm, respectively. It can be seen that at low temperature, the lifetime increases with the decrease of the dot size, thus the increase of energy. At high temperature, however, it decreases with the decrease of the dot size, and thus the increase of energy. These calculated results are in agreement *qualitatively* with our experimental results shown in Fig. 1, although the detailed comparison is difficult at present. The increase of the lifetime with the decreasing dot size at low temperature can be understood as a consequence of the penetration of the wavefunctions of electrons of the quantum dot, as observed in narrow quantum wells.[17] On the other hand, the increase of lifetime with temperature can be attributed to the thermal population of carriers, which is more effective for larger dots, as shown in Fig. 3. Therefore, we conclude that the carrier dynamics property observed in our experiments is indeed closely related to the exciton localization in the QD-like structures in our samples. The detailed calculation and its relevant equations can be found elsewhere.[18]

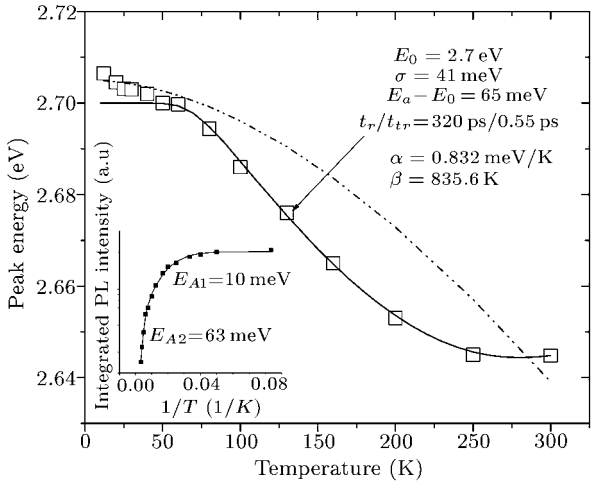

Fig. 4. Temperature dependence of the PL peak energy with the solid squares representing the experimental data and the solid line representing the fitting curve. The fitting parameters are $E_a - E_0 = 65$ meV, $\sigma = 41$ meV, and $\tau_{tr}/\tau_r = 0.55$ ps/320 ps. The inset shows an Arrhenius plot of the integrated PL intensity. The dash-dotted curve is the band gap variation with temperature for GaN with energy-shifted, and the parameters are used in the Varshni empirical formula.

Steady state PL spectra of the InGaN sample at different temperatures are also measured. Figure 4 shows the PL peak energy as a function of temperature. Obviously, it does not follow the behaviour of a semiconductor band gap predicted by the Varshni empirical formula. For comparison, the band gap variation with temperature for GaN is indicated in the figure (dash-dotted curve).[19] The observed unusual temperature variation has been previously reported in InAs QDs[20] and InGaN alloy as well.[21,22] It is often regarded as a characteristic of self-organized QDs. Here we use the thermal transfer model[8,23] to calculate the temperature dependence of peak energy in our sample. In the calculation, the distribution of the emission energy for QDs is assumed to be Gaussian-like,

$$\rho(E) \propto \exp[-(E-E_0)^2/2\sigma^2], \tag{2}$$

where E_0 and σ represent the central position of energy and the width of the distribution function, respectively. The peak energy $E(T)$ can be obtained using the equation

$$E(T) = E_0 - x(T)k_BT, \tag{3}$$

where x can be obtained by numerically solving the following equation:

$$xe^x = \left(\frac{\tau_r}{\tau_{tr}}\right)\left[\left(\frac{\sigma}{k_BT}\right)^2 - x\right]e^{(E_0-E_a)/k_BT}, \quad (4)$$

where τ_{tr} is the transfer time; τ_r is the carrier recombination time; E_a represents a barrier level that carriers must overcome to transfer. As can be seen from Fig. 4, the fitting between the calculation and the experimental data is very well when $T > 40\,\mathrm{K}$. The fitting parameters are: $E_a - E_0 = 65\,\mathrm{meV}$, $\sigma = 41\,\mathrm{meV}$, and $\tau_{tr}/\tau_r = 0.55\,\mathrm{ps}/320\,\mathrm{ps}$. The energy difference $E_a - E_0$ reflects the magnitude of the carrier localization.

The large exciton localization effect can also be obtained from the measurement of the temperature-dependent integrated PL intensity. In the inset of Fig. 4, the integrated PL intensity is shown as a function of temperature in an Arrhenius plot. It is found that the thermal quenching in our sample can be well fitted by the following equation:

$$I(t) = \frac{I_0}{1 + c_1\exp(E_{A1}/k_BT) + c_2\exp(E_{A2}/k_BT)}, \quad (5)$$

where two thermal quenching parameters are used in order to fit the dada more precisely. In other words, there are two non-radiative recombination channels involved in the PL process. From the figure, the corresponding thermal activation energies of E_{A1} and E_{A2} are extracted to be 10 and 63 meV, respectively. The value of 63 meV is in good agreement with the localization energy $E_a - E_0$ (65 meV) obtained before, implying that the observed thermal quenching of the PL is mainly due to the exciton delocalization process out of the potential barriers. The smaller activation energy of E_{A1} suggests that the exciton dissociation might play a role in the low temperature PL. Thermal release from these dissociated exciton states is accompanied by either non-radiative recombination or re-trapping on deeper states.[24]

In summary, we have studied the exciton localization effect in InGaN epilayers by using PL and TRPL. By measuring the exciton decay time as a function of the monitored emission energy at different temperatures, unusual behaviour in the energy dependence of the PL decay times is found. We have studied the effect of dot size on the radiative recombination time by numerically calculating temperature dependence of the exciton lifetime in quantum dots and found that the observed behaviour can be well explained by the exciton localization in the quantum dots. In the temperature dependent PL, the fast red shift of PL peak is observed and is well fitted by a model taking into account the thermal activation and redistribution within different potential minima in QDs. The fitted localization energy ($E_a - E_0$, 65 meV) is in good agreement with the thermal activation energy (E_{A2}, 63 meV). All these results lead to a conclusion that the exciton dynamics observed in our experiments is closely related to the exciton localization in the QD-like structures in our samples.

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