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Temperature dependence of the spontaneous emission mechanisms of localized-state heterosystem

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The temperature dependent photoluminescence (PL) spectra measured from localized-state material system is presented. Two localized-state heterosystems, including InGaN/GaN multi-quantum well (MQW) and InAs/GaAs quantum dot (QD) samples were prepared. The samples were investigated both experimentally and theoretically. It has been found that the temperature dependence of the PL peak energies from both samples behaves differently. S-shaped and anti-S-shaped PL peak energies have been observed for MQW and QD samples, respectively. We present a model which takes into account all of the key factors for the localized-carrier dynamics. The model is applied to interpret the experimental data obtained from the two kinds of material systems. Detailed discussion concerning this model provides an explicit interpretation that it is the difference in the electronic structure of the two material systems that leads to the significantly different temperature dependence of their luminescence bands.

Keywords: quantum dot, multi-quantum well, photoluminescence, localized-state.

1. Introduction

Carrier localization is a common phenomenon in many material systems such as quantum wells (QWs) and quantum dots (QDs). In QW systems, much research has been devoted to group-III nitride semiconductors [1–3]. In spite of the tremendous density of dislocations peculiar to InGaN/GaN structures grown on lattice-mismatched substrates, the high-brightness light-emitting diodes and cw blue laser diodes based on InGaN structures have been obtained with high performance and high quantum efficiency [4]. Recombination mechanisms in InGaN/GaN multi-quantum wells (MQWs) have been extensively studied and attributed to the emission from localized band-tail states originating from QD-like and phase-separated indium-rich regions in the wells [5, 6]. This carrier localization formed in the plane of the layers enhances the quantum efficiency by suppressing lateral carrier diffusion, thereby reducing the probability of carriers entering nonradiative recombination centers.

In self-assembled InAs/GaAs QD systems, the strong localization of the electronic wave function leads to an atom-like electron density of states and can be used for the realization of novel optoelectronic devices like QD lasers [7–9]. The unusual variations of the emission energy of the luminescence bands in InAs self-assembled QDs have been investigated extensively [10–13]. A fast redshift of the emission wavelength is usually observed in the mid-temperature range. The feature has been explained by enhanced carrier redistribution among dots due to the thermal escape of carriers having higher energies and their subsequent recapture by dots emitting on the low-energy side of the distribution.

The InGaN alloys with indium-rich clusters are very similar to the InAs/GaAs QD systems. However, the temperature dependent behaviors of luminescence from these two material systems are quite different. With increasing temperatures, the luminescence peaks exhibit a blueshift for InGaN MQWs but a quick redshift for InAs QDs. In order to get a better understanding of the spontaneous emission mechanisms of the localized-state ensemble system, we present a model in the article to describe the anomalous spectral features of InGaN MQWs and InAs QDs. All the relevant carrier thermalization and quenching processes active in localized-state system are taken into account. The model is applied to interpret the experimental data obtained from the MQWs and QDs material systems. From quantitative analysis, the physical origins that cause the different thermal behaviors of the samples are demonstrated.

2. Experimental details

In the present study, the InGaN/GaN MQWs and the self-assembled InAs/GaAs QD samples were both created using metal-organic chemical vapor epitaxy system. The InGaN/GaN MQW sample was grown on *c*-plane sapphire substrates. The layer structure of the sample consisted of a 20-nm-thick GaN buffer layer, a 3-μm-thick Si-doped *n*-type GaN layer, an undoped GaN layer possessing five periods of $In_{0.18}Ga_{0.82}N$/GaN MQWs, and a 100-nm-thick Mg-doped *p*-type GaN layer. The thicknesses of the InGaN wells and the GaN barriers in the MQW structure were 2 nm and 11 nm, respectively. The temperature dependent photoluminescence (PL) spectra were measured using a He-Cd laser operating at a wavelength of 325 nm and the average excitation intensity was 20 mW. The sample was mounted on a Cu cold stage where the temperature T was varied from 20 to 300 K. The luminescence signal was dispersed through a 0.5 m monochromator and detected by using a Si photodiode by standard lock-in amplification technique. The InAs/GaAs QD sample was grown on (100) 2°-tilted toward (111)A GaAs substrate. The heterostructure included a 400-nm Si-doped GaAs buffer layer, an InAs QD active region of 3.3 monolayers, and a 100-nm undoped GaAs capping layer. The temperature dependent PL spectra were measured using the same He-Cd laser. The sample was mounted in a closed-cycle He cryostat, which allows measurements in a temperature range from 20 to 300 K. The luminescence signal was detected using a Ge photodiode.

3. Results and discussion

Figure 1 shows the temperature dependence of the PL peak position over a temperature range from 20 to 300 K of InGaN/GaN MQW sample and InAs/GaAs QD sample. It is obvious that the luminescence bands of MQW and QD samples exhibit quite different temperature dependence. For the MQW sample as shown in Fig. 1**a**, a well-established S-shaped temperature behavior is observed as the temperature increases. The peak energy redshifts at temperature lower than 100 K; then it blueshifts in the range of up to 220 K and redshifts again afterwards. The "S-shaped" temperature dependence of the luminescence peak energy is a fingerprint of the existence of localization effect [5, 14, 15]. For the QD sample shown in Fig. 1**b**, a quick redshift of emission energy with increasing temperatures is observed. From 20 to 200 K, a redshift of 45.4 meV is obtained for the sample. It can be seen that the redshift is larger than the InAs bulk band gap which is calculated by the Varshni law (~35.8 meV) [16, 17]:

$$E_g(T) = E_g(0) - \frac{\alpha T^2}{T + \beta} \tag{1}$$

where $E_g(T)$ is the InAs bulk emission energy at T, $E_g(0)$ is the energy gap at 0 K, and α and β are Varshni's fitting parameters. The anomalous fast redshift of luminescence energy of QD sample has been attributed to a thermally enhanced carrier redistribution among dots that is caused by carrier thermionic emission through the wetting layer [12, 13]. At low temperature, carriers are captured randomly into the QDs. With elevated temperature, carriers may be thermally activated outside the small dots into the wetting layer and then preferentially relaxed into large dots with relatively low-localized energy states. Thus faster peak energy decrease is observed than that of the InAs bulk band gap.

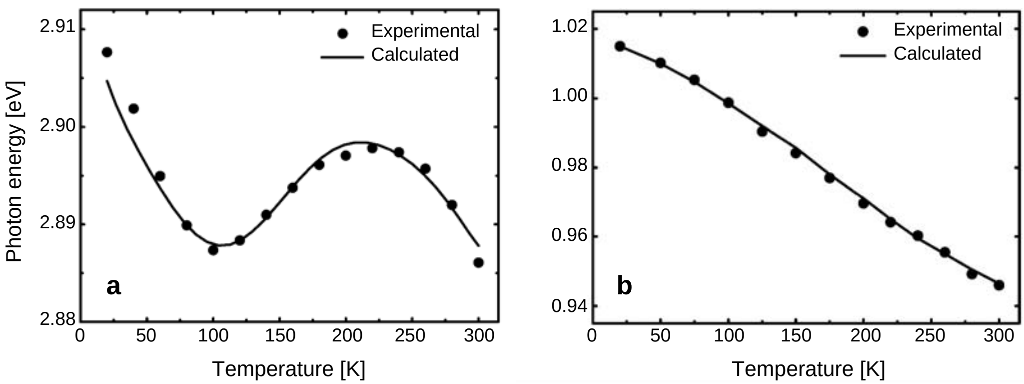

Fig. 1. Temperature dependence of peak position of PL spectra from InGaN/GaN MQW sample (**a**), and InAs QD sample (**b**). The solid lines are fitting curves based on the rate-equation model with fitting parameters listed in the Table.

Both of the InAs QD sample and the InGaN MQW sample in this work exhibit the carrier localization effect on their PL spectra. It can be expected that there are similar mechanisms of carrier transferring. To model the temperature dependent behaviors of the emission peaks of PL spectra for the samples, we use a rate-equation model to describe the carrier dynamics in the system with localized states. Suppose the energetic distribution of the localized states is a Gaussian-like distribution function $n(E)$ of energy, and $n_f(E)$ and $n_e(E)$ are the numbers of filled and empty localized states, respectively:

$$n(E) \propto \exp\left[-\frac{(E-E_0)^2}{2\sigma^2}\right] \tag{2}$$

$$n(E) = n_f(E) + n_e(E) \tag{3}$$

where E_0 is the peak energy position and σ is the standard deviation of the distribution.

The carriers are injected into the localized-state system at a constant rate g by an external excitation source. Carriers in localized state are assumed to be radiatively recombined at rate γ_r, or thermally escape to an energetic position E_a when the temperature is sufficiently high. The energy level E_a represents the energy level that the carriers must overcome to transfer, and the coefficient corresponding to escape from the localized state E to E_a is given as follows:

$$\gamma_{\text{esc}} = \gamma_0 \exp\left(-\frac{E_a - E}{k_B T}\right) \tag{4}$$

where γ_0 is the intrinsic escape coefficient and k_B is the Boltzmann constant. The state filling effect is significant in the QD system because of the reduced density of states; it should be taken into account [11, 18]. This effect implies that the capturing and thermal escaping processes in the system are proportional to the number of empty states into which the carriers arrive. Thus, we can write a set of rate equations that describe the rate of change of carriers in the localized states:

$$\frac{\mathrm{d}n_r}{\mathrm{d}t} = g - n_r\gamma_{\text{nr}} - \int n_r\gamma_c n_e(E)\mathrm{d}E + \int n_f(E)\gamma_{\text{esc}}\mathrm{d}E \tag{5}$$

$$\frac{\mathrm{d}n_f(E)}{\mathrm{d}t} = -n_f(E)\gamma_{\text{esc}} - n_f(E)\gamma_r + n_r\gamma_c n_e(E) \tag{6}$$

where n_r is the number of carriers in E_a, γ_{nr} – the nonradiative recombination rate coefficient of n_r, γ_c – the capture rate of carriers by the localized states. The coupled rate equations are solved numerically under a steady state assumption by fitting the integrated PL intensity. Once the carrier distribution function $n_f(E)$ is determined, the luminescence spectrum $I(E)$ is expressed as

$$I(E) \propto n_f(E)\gamma_r \tag{7}$$

From Equation (7), the peak energies of the spectra for the samples are obtained, denoted as $E_{\text{loc}}(T)$. The luminescence spectra $I(E)$ and the peak energy $E_{\text{loc}}(T)$ are only considering the thermal redistribution effect of carriers within the localized states. The band gap of semiconductor is itself temperature dependent and is described by the Varshni law. After taking into account this factor, the temperature dependent peak energies of the samples are determined as

$$E_{\text{peak}}(T) = E_{\text{loc}}(T) - \frac{\alpha T^2}{T+\beta} \tag{8}$$

For InGaN MQW sample, $E_0 = 2.993$ eV, $E_a = 2.913$ eV and $\sigma = 34$ meV are adopted in modeling the experimental results. For InAs QD sample, the density of states is assumed to be proportional to the Gaussian distributions. The parameters $E_0 = 1.014$ eV, $E_a = 1.3$ eV, and $\sigma = 12.3$ meV are chosen to match the peak energies and linewidths of the lowest temperature PL spectra [12, 13]. The fitting curves of temperature dependent PL peak energies for both samples based on the model described above are also shown in Fig. 1 and the fitting parameters are listed in the Table. Despite the simplicity of this model, the calculated values are in good agreement with the experimental data, demonstrating that the model is indeed realistic.

T a b l e. Fitting parameters used in the model for calculating the temperature dependent luminescence spectra for InGaN/GaN MQW sample and InAs QD sample.

	γ_0 [s^{-1}]	γ_{nr} [s^{-1}]	γ_c [cm^{2}s^{-1}]	γ_r [s^{-1}]
InGaN MQW sample	5×10^{17}	6×10^{16}	10^{12}	10^9
InGaN QD sample	1×10^{19}	1×10^{17}	6.7×10^9	2×10^9

To investigate the difference of temperature dependent behaviors between the InGaN MQW and InAs QD samples, it is important to discuss further the energy level E_a. Schematic diagrams of electronic pictures for MQW sample and QD sample are shown in Fig. 2**a** and Fig. 2**b**, respectively. The magnitude of $E_a - E_0$ is a measure of thermal activation energy. In QD system, the value of $E_a - E_0$ is 0.326 eV, the thermal activation energy locates at an energy 326 meV above the central position of the localized states. The origin of such an energy level comes from the presence of a wetting layer in self-assembled QD structures. As the temperature increases, carriers located at shallow potential minima can be thermally activated and the carriers migrate to the wetting layer, being then retrapped by QDs. Thus, the peak energy experiences

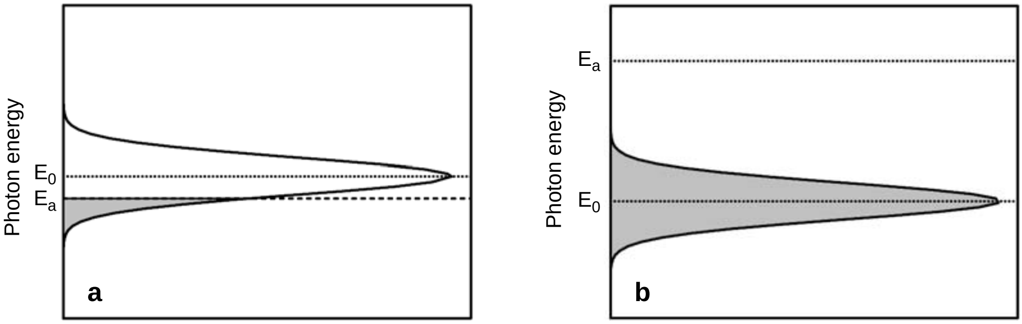

Fig. 2. Schematic diagrams of electronic structures for InGaN MQW sample (**a**), and InAs QD sample (**b**).

a redshift because the emitting QDs are characterized by energetically lower states. For MQW sample, the value of $E_a - E_0$ is -0.08 eV, and is located at 80 meV below the central energy position of the localized-state distribution. The increasing temperatures enable carriers to populate the higher levels of the localized state, resulting in a blueshift of the peak energy. In the high temperature range, the localization effect is compensated and overcome by the redshift of peak energy due to the temperature shrinkage of band gap energy, leading to a redshift of peak energy.

The anomalous temperature dependent S-shaped and anti-S-shaped peak energies for MQW and QD samples, respectively, are both demonstrated by the model. It is obvious that E_a relative to the central energy of the density of states for the localized carriers has a decisive influence on the temperature dependence of the luminescence peak position. From the physical viewpoint, E_a can be considered as the energetic level below which all the localized states are occupied by carriers at 0 K, like the Fermi level in the Fermi–Dirac distribution function [19]. To further analyze the carrier dynamics in the localized-state system, the simulated temperature dependent $I(E)$ and $E_{\text{loc}}(T)$, considering the thermal redistribution effect for the MQW sample, are shown in Fig. 3**a** and Fig. 3**b**, respectively. The emission energies which are calculated by the Varshni law and the peak energies $E_{\text{peak}}(T)$ which combine both thermal redistribution effect and band gap shrinkage for MQW sample are also shown in Fig. 3**b**. At low temperatures, carriers exist in the localized states with lower energies. As the temperature slightly increases, weakly localized carriers are thermally activated and redistributed into other strongly localized states, as shown in Fig. 3**a**. It is consistent with the redshift of energies $E_{\text{loc}}(T)$ and the induced quick redshift of energies $E_{\text{peak}}(T)$ in the low temperature range, which can be seen in Fig. 3**b**. After the effect of redistribution is saturated, the increasing temperatures enable carriers to achieve thermal equilibrium and to populate the higher levels of the localized state, thus resulting in a blueshift of the peak energy $E_{\text{loc}}(T)$. An almost constant $I(E)$ linewidth in the temperature range $T > 140$ K indicates an occurrence of thermal equilibrium energy distribution. The blueshift of $E_{\text{peak}}(T)$ slows down in the high

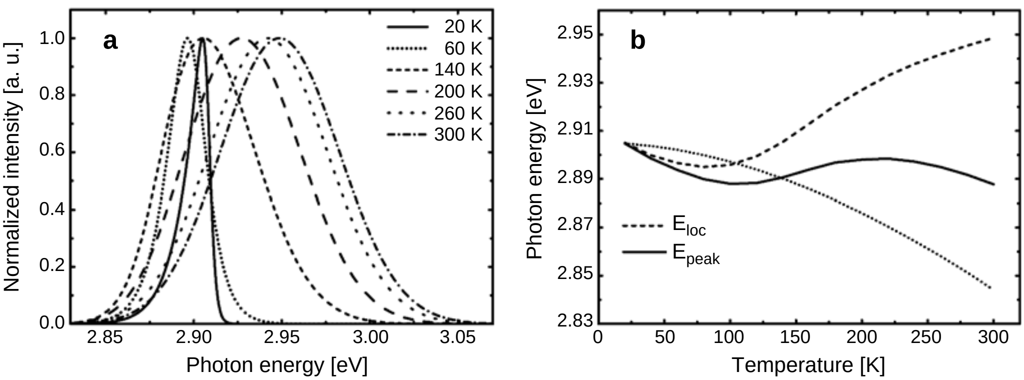

Fig. 3. The simulated luminescence spectra $I(E)$ (**a**), and emission energies $E_{\mathrm{loc}}(T)$ and $E_{\mathrm{peak}}(T)$ under various temperatures for InGaN MQW sample; the dotted line represents the results calculated by the Varshni law (**b**).

temperature range and is overcome by the band gap shrinkage, resulting in a redshift of PL peak energies, as shown in Fig. 3**b**.

The corresponding plots for InAs QD sample are shown in Fig. 4. In InAs QD sample, most of the localized states are occupied by the injected carriers at low temperature. Referring to the calculated spectra $I(E)$ shown in Fig. 4**a**, the shape of $I(E)$ is almost unchanged as $T < 160$ K, resulting in the fixed energy $E_{\mathrm{loc}}(T)$ shown in Fig. 4**b**. In the temperature range 180 K–260 K, it is clearly seen from the simulated $I(E)$ spectra that the carriers distributed on the higher-energy side are decreasing. As the temperature increases, carriers with higher energies begin to escape to the wetting layer. Some of them are retrapped by the QDs, giving rise to thermal redistribution. The thermal redistribution of carriers among QDs is evident, inducing a redshift of

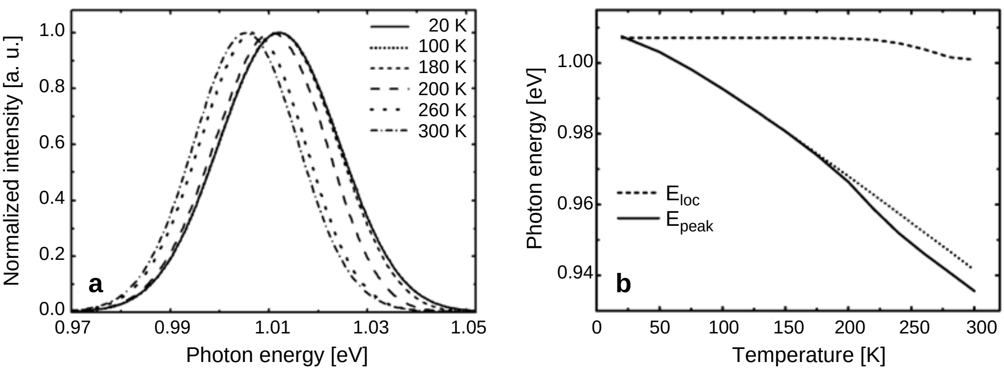

Fig. 4. The simulated luminescence spectra $I(E)$ (**a**), and emission energies $E_{\mathrm{loc}}(T)$ and $E_{\mathrm{peak}}(T)$ under various temperatures for InAs QD sample; the dotted line represents the results calculated by the Varshni law (**b**).

peak energy $E_{loc}(T)$. As the temperature is higher than 260 K, the thermal redistribution effect is gradually saturated and the redshift of $E_{loc}(T)$ and $E_{peak}(T)$ retards, in accordance with data measured in our experiments.

4. Conclusions

In this paper, the InGaN/GaN MQW sample and the InAs/GaAs self-assembled QD sample were grown and the PL spectra under different temperatures were measured. It has been found that the temperature dependence of the luminescence bands of the two samples is quite different. We study the optical properties of these two localized-state material systems based on a rate-equation model. All of the key factors for the localized-carrier dynamics are considered in our model. The anomalous temperature dependent S-shaped and anti-S-shaped peak energy of MQW and QD samples, respectively, are both demonstrated by the model. Furthermore, the detailed discussion of the simulated PL lineshape and the emission energies under various temperatures are derived to investigate the carrier dynamics of the localized-state heterosystem. Our study provides an explicit interpretation to explain the significantly different temperature dependence of luminescence bands for the two material systems. It is concluded that the model is useful for understanding the emission mechanisms of carriers in localized-state material systems.

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