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Quantum-confinement effect on recombination dynamics and carrier localization in cubic InN and $In_xGa_{1-x}N$ quantum boxes

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ABSTRACT

In this study, the quantum confinement effect on recombination dynamics and carrier localization in cubic InN (*c*-InN) and cubic $In_xGa_{1-x}N$ (*c*-$In_xGa_{1-x}N$) low dimensional structures are theoretically examined. The small InN and In-rich $In_xGa_{1-x}N$ low dimensional structures show a strong quantum confinement effect, which results in ground states away from the band edge and discrete eigen-states. Depending on composition, temperature, and size of the InN and $In_xGa_{1-x}N$ low dimensional structures, quantum confinement effect can affect the exciton dimensions (D). In InN quantum cubes, the strong quantum confinement effect leads to temperature-dependent radiative lifetimes showing a large size variation. The nearly-temperature-independent and shorter radiative lifetimes in small InN and In-rich $In_xGa_{1-x}N$ low dimensional structures suggest that the strong quantum confinement leads to *0* D carrier confinement, stronger carrier localization, and high recombination efficiency. Reported radiative lifetimes were found to match very well with our simulation results of In-rich quantum cubes, which indicates that spontaneous emissions come from the radiative recombination of localized excitons in In-rich $In_xGa_{1-x}N$ clusters. The simulation results could provide important information for the designs and interpretations of *c*-InN and *c*-$In_xGa_{1-x}N$ devices.

1. Introduction

InN and In-rich $In_xGa_{1-x}N$ compounds have been a subject of intense study due to their promising properties for optoelectronics, photovoltaic, and high-speed electronics applications [1–4]. Cubic InN (*c*-InN) shows superior transport properties and the near absence of polarization-induced electrostatic field [1–3]. There have been reports of the growth of metastable *c*-InN on SiC and sapphire substrates [2,3] and cubic InGaN on GaAs substrates [4]. Synthesis of InN nanostructures, such as quantum dots (QDs), represents a unique approach to ascertaining their intrinsic properties [5–7]. InN quasi-QDs generally exists in wurtzite structures (hexagonal, *h*-InN) as the thermodynamically stable phase. In anticipation of the increasing interest in InN nanostructures, we aim to report a theoretical investigation regarding the electronic and recombination properties that would enable the design and interpretations of *c*-InN and cubic InGaN (*c*-InGaN) low dimensional structures. Furthermore, in our previous study, some phenomena attributed to the enhanced quantum confinement effect in low dimensional nanostructures were observed: (a) the ground states shift upward; (b) the number of eigen-states is decreased; (c) the eigen-energies are discrete [8].

Creation of artificial nanostructures with controlled dimensionality can test our model of exciton behaviors. Excitons confined in *n*-dimensions (*n*-D excitons) have a radiative lifetime with temperature dependence of $T^{n/2}$ [9]. The low miscibility of InN and GaN leads to the formation of In-rich $In_xGa_{1-x}N$ clusters [10,11]. The In-rich $In_xGa_{1-x}N$ clusters act as quasi-QDs and provide local potential minimums that suppress the diffusion of carriers toward nonradiative defects. It was claimed that the electroluminescence (EL) emissions come from the recombination of localized excitons in In-rich $In_xGa_{1-x}N$ clusters [12–14]. For *c*-InGaN clusters in *c*-InGaN/GaN heterostructures, it was found that the excition dimension changes from *0* D (at low temperature) to *2* D (at high temperature) [15]. Although carrier localization in In-rich *c*-InGaN clusters was proposed to explain the optical properties of *c*-InGaN/GaN heterostructures [15], the impact of the quantum confinement effect on the recombination dynamics and carrier localization in *c*-InN and *c*-$In_xGa_{1-x}N$ low dimensional structures has not been closely investigated.

In this study, the quantum confinement effect on the recombination dynamics and carrier localization in *c*-InN and *c*-$In_xGa_{1-x}N$ low dimensional structures is theoretically examined. Depending on the composition, temperature, and size of InN and $In_xGa_{1-x}N$ low dimensional structures, the quantum confinement effect can affect the exciton dimensions. The nearly temperature-independent and shorter radiative lifetimes in small InN and In-rich $In_xGa_{1-x}N$ low dimensional structures suggest that the strong quantum confinement leads to *0* D carrier confinement, stronger carrier localization, and high recombination efficiency. Reported radiative lifetimes were found to match very well with our simulation results using In-rich quantum cubes, which

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indicates that spontaneous emissions come from the radiative recombination of localized excitons in In-rich $In_xGa_{1-x}N$ clusters.

This paper is organized as follows: In section 2, theoretical calculations of the band structures of $In_xGa_{1-x}N$/GaN quantum cubes are described. In section 3, the quantum-confinement effect on the recombination dynamics in *c*-InN and *c*-$In_xGa_{1-x}N$ low dimensional structures are discussed. In section 4, carrier localization in *c*-InN and *c*-$In_xGa_{1-x}N$ low dimensional structures are discussed. Finally, conclusions are drawn in section 5.

2. Theoretical calculations

A series expansion model was conducted to calculate the band structures of size- and composition-dependent *c*-$In_xGa_{1-x}N$ quantum cubes ($L_x=L_y=L_z=L$) embedded in GaN matrixes [17,18]. The hydrostatic strain was thought to modify the band structure [16]. The strain was assumed to be compressive inside the quantum cube and zero outside the quantum cube. The eigen-valued problem was solved with a 10×10×10 tensor for either conduction or valence bands . The details of the calculations and physical parameters were described in our previous study [8].

The radiative recombination lifetime was calculated. At zero temperature (0 K), electrons and holes relax to the ground state and the radiative recombination lifetime $\tau(0\ K;\ x)$ for *c*-$In_xGa_{1-x}N$ is determined by only the ground state [17]:

$$\tau(0\,K;x)=\frac{\pi\varepsilon_0\hbar^2c^3m_0^2(x)}{n(x)e^2E^e_{hh(lh)}(0\,K;x)|E_P(x)|^2\left|\sum\limits_{\substack{l_e=l_h\\ m_e=m_h\\ n_e=n_h}} D^0_{l_e,m_e,n_e,l_h,m_h,n_h}(0\,K;x)\right|^2}=\frac{\tau_c(0\,K;x)}{\left|\sum\limits_{\substack{l_e=l_h\\ m_e=m_h\\ n_e=n_h}} D^0_{l_e,m_e,n_e,l_h,m_h,n_h}(0\,K;x)\right|^2}\qquad(1)$$

where $|E_P(x)|^2$ is the squared momentum matrix element; and ε_0, c, $m_0(x)$, and $n(x)$ are the dielectric constant, the velocity of light, the free electron mass, and the refractive index of the material, respectively. $E^e_{hh(lh)}(0\ K;\ x)$ are the transition energies from electron to heavy-hole (light-hole) eigenstates; $D^0_{l_e,m_e,n_e,l_h,m_h,n_h}(0\ K;\ x)$ and $|D^0_{l_e,m_e,n_e,l_h,m_h,n_h}(0\ K;\ x)|^2$ are the expansion coefficient and oscillator strength of ground-state electron-hole wavefunction, respectively. The composition-dependent $|E_P(x)|^2$, $m_0(x)$, and $n(x)$ for *c*-$In_xGa_{1-x}N$ were expressed as the linear interpolation formula of InN and GaN [8].

To calculate the temperature- and composition-dependent recombination lifetime, $\tau(T;\ x)$, the parameters used in Eq. (1) should be temperature-dependent. Because the temperature dependence of some parameters are still unknown, we only consider the dependence of transition energies, $E^e_{hh(lh)}(T;x)$, and oscillator strength, $|D^{av}(T;\ x)|^2$. The temperature- and composition-dependent transition energy, $E^e_{hh(lh)}(T;\ x)$, between the ground electron state and ground heavy-hole (light-hole) state is defined as:

$$E^e_{hh(lh)}(T;x)\equiv E^c_0(0;x)+\delta E_c(0;x)+E_g(T;x)+\delta E_v(0;x)+E_0^{hh(lh)}(0;x)\qquad(2)$$

where $E^c_0(0;\ x)$, $E_0{}^{hh}(0;\ x)$, and $E^{lh}_0(0;\ x)$ are the electron, heavy-hole, and light-hole eigen-energies of the ground states at 0 K. $\delta E_c(0;\ x)$ and $\delta E_v(0;\ x)$ are the hydrostatic-strain-induced energy shifts for the conduction band and valence band at 0 K, respectively. The temperature- and composition-dependent bandgap energy $E_g^{In_xGa_{1-x}N}(T;\ x)$ can be expressed as:

$$E_g^{In_xGa_{1-x}N}(T;x)=xE_g^{InN}(T)+(1-x)E_g^{GaN}(T)-bx(1-x)\qquad(3)$$

where $E_g^{InN}(T)$ *and* $E_g^{GaN}(T)$ are the temperature-dependent band-gap energies of InN and GaN, respectively. $b(=1.4\ eV)$ is a bowing constant. $E_g^{InN(GaN)}(T)$, are assumed to obey Varshni' equation:

$$E_g^{InN(GaN)}(T)=E_g^{InN(GaN)}(0K)-\frac{\alpha T^2}{T+\beta}\qquad(4)$$

where α and β are the fitting parameters. α and β are 0.593 meV/K and 600 K for GaN, respectively, while they are 0.245 meV/K and 624 K for InN [19].

In addition, at higher temperature(s), thermal energy helps carriers to distribute to higher excited levels. The carrier density is assumed to be lower than box density and carrier relaxation time is quicker than carrier recombination. The Boltzman distribution was used to average the oscillator strengths of the ground and higher excited states. The averaged oscillator strength $|D^{av}(T;\ x)|^2$ is given by [17]:

$$|D^{av}(T;x)|^2=\frac{\sum\limits_i\left|\sum\limits_{\substack{l_e=l_h\\ m_e=m_h\\ n_e=n_h}} D^i_{l_e,m_e,n_e,l_h,m_h,n_h}(0K;x)\right|^2 e^{-E_i(0K;x)/k_BT}}{\sum\limits_i e^{-E_i(0K;x)/k_BT}}\qquad(5)$$

where k_BT is the thermal energy. $E_i(0\ K;\ x)$ and $|D^0_{l_e,m_e,n_e,l_h,m_h,n_h}(0\ K;\ x)|^2$ are the i-th eigen-energy and oscillator strength of the electron-hole wavefunction, respectively. It is noted that averaged oscillator strength was calculated by using the eigen-energy at 0 K.

Therefore, the temperature- and composition-dependent recombination lifetime, $\tau(T;\ x)$, is calculated as [17]:

$$\tau(T;x)=\frac{\pi\varepsilon_0\hbar^2c^3m_0^2(x)}{n(x)e^2E^e_{hh(lh)}(T;x)|E_P(x)|^2|D^{av}(T;x)|^2}=\frac{\tau_c(T;x)}{|D^{av}(T;x)|^2}.\qquad(6)$$

For n-D excitons, the temperature-dependent oscillator strength depends on temperature, as $(k_BT)^{-n/2}$. The radiative recombination

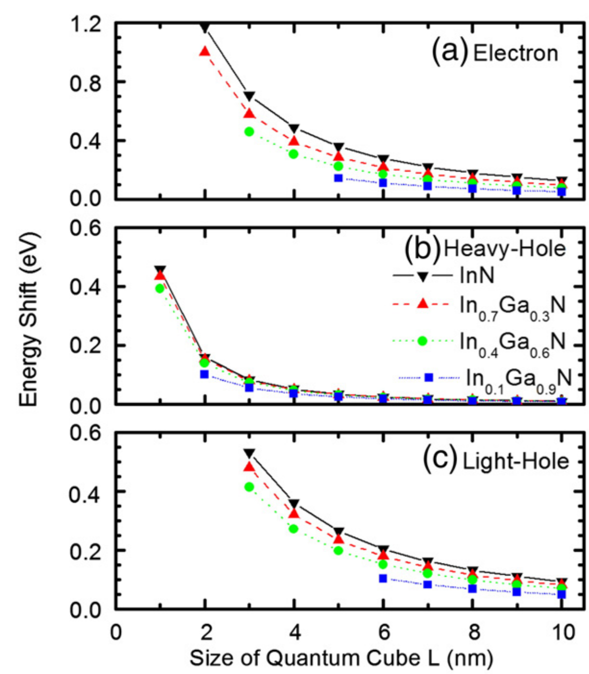

Fig. 1. (a) Electron ground-state energy, $E^c_0(0\ K)$, (b) heavy-hole ground-state energy, $E^{hh}_0(0\ K)$, and (c) light-hole ground-state energy, $E^{lh}_0(0\ K)$, as functions of quantum cube size for $In_xGa_{1-x}N$ quantum cubes at 0 K.

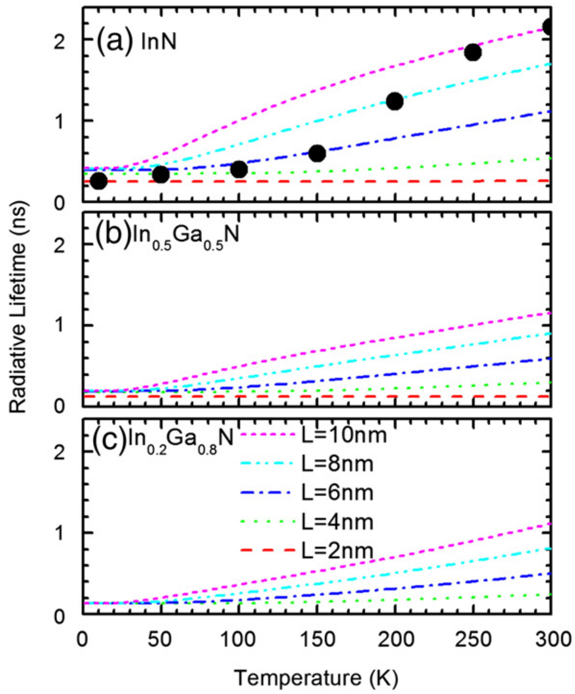

Fig. 2. Temperature dependence of radiative recombination lifetimes, $\tau^e_{hh}(T;\ x)$, for (a) InN, (b) $In_{0.5}Ga_{0.5}N$, and (c) $In_{0.2}Ga_{0.8}N$ quantum cubes. Transitions are between electrons and heavy-holes eigen-states. For comparison, the reported experimental data were plotted as filled circles (●) in Fig. 2 (a).

lifetime is inversely proportional to oscillator strength. Hence, for *n*-D excitons, the radiative recombination lifetime is dependent on temperature, as $(k_BT)^{n/2}$ [3,4].

3. Quantum confinement effect on recombination dynamics in *c*-InN and *c*-$In_xGa_{1-x}N$ low dimensional structures

Fig. 1(a), (b), and (c) shows the electron eigen-energies $E_0^c(0\ K)$, heavy-hole eigen-energies $E_0^{hh}(0\ K)$, and light-hole eigen-energies $E_0^{lh}(0\ K)$ of the ground states as functions of quantum cube size *L* at 0 K, respectively. As In composition increases, $E_0^c(0\ K)$, $E_0^{hh}(0\ K)$ and $E_0^{lh}(0\ K)$ become larger in smaller cubes. In 3 nm InN quantum cubes, $E_0^c(0\ K)$, $E_0^{hh}(0\ K)$ and $E_0^{lh}(0\ K)$ can be as large as 0.708 eV, 0.083 eV, and 0.531 eV, respectively. These imply that the quantum confinement effect becomes dominant in determining the optical properties of smaller InN and $In_xGa_{1-x}N$ low dimensional structures. Besides, because the effective mass of the heavy hole is larger than those of electron and light hole, $E_0^{hh}(0\ K)$ are smaller than $E_0^c(0\ K)$ and $E_0^{lh}(0\ K)$.

Fig. 2(a), (b), and (c) shows the temperature dependence of the radiative recombination lifetime, $\tau^e_{hh}(T;\ x)$, for InN, $In_{0.5}Ga_{0.5}N$, and $In_{0.2}Ga_{0.8}N$ quantum cubes, respectively. Transitions between electron and heavy-hole eigen-states are considered. $\tau^e_{hh}(T;\ x)$ remain constant at low temperatures while $\tau^e_{hh}(T;\ x)$ are approximately proportional to temperature as $\sim T^{3/2}$ at high temperatures. This implies that $\tau^e_{hh}(T;\ x)$ shows hybrid features of *0* D and quasi-*3* D as a function of temperature [13,15]. At low temperatures, $\tau^e_{hh}(T)$ remain constant which indicates very tight confinement in a *0* D system. The effect of quantum confinement is manifested in the spacing of eigen-states; smaller cubes (small *L*), higher indium compositions, and lighter effective masses (for electrons and light holes) tend to enhance the *0* D characteristics. Meanwhile, $\tau^e_{hh}(T;\ x)$ for larger quantum cubes are approximately proportional to temperature as $\sim T^{3/2}$ at high temperatures. A gradual increase of $\tau^e_{hh}(T;\ x)$ toward high temperature is a result of reduced electron-hole overlap. Our calculation indicates that the thermalization of heavy-holes into the higher, closely-spaced levels is responsible for the higher-dimensional behavior. It is noted that although the quasi-3 D behavior of $\tau^e_{hh}(T;\ x)$ in larger quantum boxes at higher temperature(s) are observed, the excitons are still localized in the quantum boxes. In addition, due to the strong quantum confinement effect and small band gap of InN, $\tau^e_{hh}(T;\ x)$ displays pronounced variations with respect to the size of quantum cubes. According to Eq. (1), this leads to larger values and large size variations of τ_c. In small cubes, $\tau^e_{hh}(T;\ x)$ are nearly constant and show higher transition temperatures between *0* D and *3* D. The trends suggest that the strong quantum confinement in small InN and $In_xGa_{1-x}N$ low dimensional structures leads to a *0* D carrier confinement and a very short radiative lifetime.

For comparison, the reported radiative lifetimes of a cubic $In_{0.1}Ga_{0.9}N$/GaN multiple quantum wells sample are plotted as filled circles (●) in Fig. 2(a) [15]. The experimental radiative lifetimes show features of strong quantum confinement effect and QD-like *0* D behavior below 100 K. While the measured radiative lifetimes fall in the range of our calculation, a detailed fitting and agreement could not be attained at present since there is no provision in our modeling to account for the statistical fluctuations of the $In_xGa_{1-x}N$ QDs in size and composition. At high temperature, because thermal distributions of excited states in larger QDs are more efficient than those in smaller QDs, the inverse oscillation strengths in larger QDs are larger (not shown here). Hence, the observed radiative lifetimes qualitatively show the behaviors of larger QDs. Furthermore, the experimental radiative lifetimes can nearly match the trend of In-rich quantum cubes in our simulation results. This trend clearly indicates that spontaneous emissions come from the radiative recombination of localized excitons in In-rich $In_xGa_{1-x}N$ clusters. In general, the recombination lifetimes in *c*-$In_xGa_{1-x}N$ are shorter than those of *h*-$In_xGa_{1-x}N$ with similar In composition [13,15]. It is possible that in the absence of the internal electrostatic fields, the recombination efficiency of *c*-$In_xGa_{1-x}N$ may be intrinsically superior to that of *h*-$In_xGa_{1-x}N$.

Fig. 3 shows the temperature-dependent radiative recombination lifetimes, $\tau^e_{lh}(T;\ x)$, for transition from electron to light-hole eigen-states. The trends of temperature-dependent $\tau^e_{lh}(T;\ x)$ show a feature

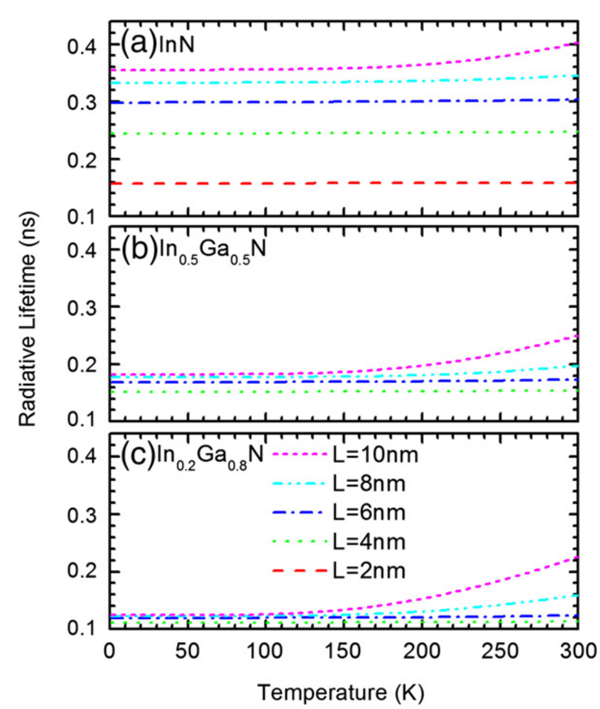

Fig. 3. Temperature dependence of radiative recombination lifetimes, $\tau^e_{lh}(T;\ x)$, for (a) InN, (b) $In_{0.5}Ga_{0.5}N$, and (c) $In_{0.2}Ga_{0.8}N$ quantum cubes. Transitions are between electrons and light-holes eigen-states.

of excition dimensions between *0* D and *3* D. Because of the strong quantum confinement effect in InN quantum cubes, some observed features of $\tau^e_{lh}(T;\,x)$ are: (1) dimension transitions at high temperatures, (2) nearly temperature-independent constants, and (3) larger size variations. Also, compared with $\tau^e_{hh}(T;\,x)$, $\tau^e_{lh}(T;\,x)$ show shorter radiative recombination lifetimes and dimension transitions at high temperatures. The enhanced quantum confinement effect and discrete eigen-energies E^{lh}_n of light holes can also explain the trends. Those trends also imply that electron transitions from electron to light-hole eigen-states in InN and In-rich $In_xGa_{1-x}N$ low dimensional structures leads to high recombination efficiency. It is possible that the use of compressive-strain engineering to alter the valence band edge may preferentially facilitate the filling of the light-hole band and exploit the efficacy in radiative recombination [20].

4. Carrier localization in *c*-InN and *c*-$In_xGa_{1-x}N$ low dimensional structures

In order to investigate the thermal effect on dimension transition, the transition temperatures are plotted in Fig. 4. Because of the enhanced quantum confinement effect, an increasing trend of transition temperatures in smaller cubes was observed. The transition temperature roughly corresponds to the energy for the thermal population (E_{300K}~26 *meV*). The thermal energy is approximately equal to the energy spacing between the ground state and the first heavy-hole excited state. The result is consistent with the previous argument that the thermalization of heavy-hole into the higher, closely-spaced levels is responsible for the higher-dimensional behavior. In addition, the higher transition temperature for the transition from electron to light-hole eigen-states can be due to the discrete eigen-states and tight confinement effect of light-holes. Because of the strong confinement effect in smaller QDs, some transitions are beyond room temperature and not shown here.

Fig. 5(a) shows the radiative lifetimes, $\tau^e_{hh}(0\ K;\,x)$, of $In_xGa_{1-x}N$ cubes at 0 K for electron transitions from electron to heavy-hole eigen-states. $\tau^e_{hh}(0\ K;\,x)$ show an increasing trend and a larger size variation in In-rich $In_xGa_{1-x}N$ quantum cubes. This trend is typical of carrier localization in our previous study [21]. We speculate that the quantum confinement effect can enhance carrier localization in In-rich $In_xGa_{1-x}N$ clusters. Because the strong quantum confinement effect and small band gap of InN leads to a large size variation of calculated electron transition energies E^e_{hh}, $\tau^e_{hh}(0\ K;\,x)$ shows a large size variation. In addition, the shorter $\tau^e_{hh}(0\ K;\,x)$ in smaller InN and In-rich $In_xGa_{1-x}N$ quantum cubes implies that the quantum confinement effect can also leads to better recombination efficiency. On the other hand, Fig. 5(b) shows the

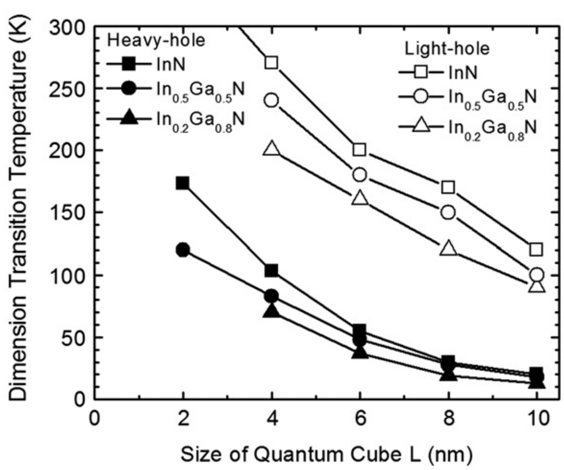

Fig. 4. Dimension transition temperature as functions of quantum cube size for transition from electron to heavy-hole and light-hole eigen-states.

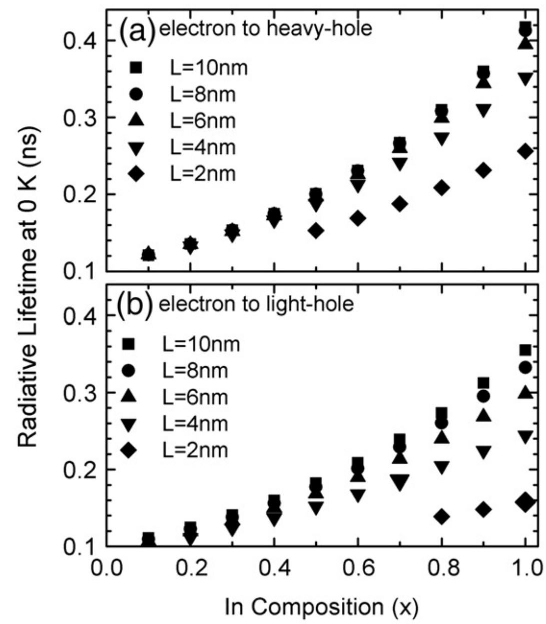

Fig. 5. (a) Radiative lifetime, $\tau^e_{hh}(0\ K;\,x)$, as functions of indium composition at 0 K for transitions from electron to heavy-hole eigen-states. (b) Radiative lifetime, $\tau^e_{lh}(0\ K;\,x)$, as functions of indium composition at 0 K for transitions from electron to light-hole eigen-states.

radiative lifetimes, $\tau^e_{lh}(0\ K;\,x)$, of $In_xGa_{1-x}N$ cubes at 0 K for electron transition energies, E^e_{lh}, from electrons to light-hole eigen-states. The trends of $\tau^e_{lh}(0\ K;\,x)$ are similar to those of $\tau^e_{hh}(T=0\ K)$. Because the enhanced quantum confinement effect of the light holes leads to larger values and larger size variations of E^e_{lh}, $\tau^e_{lh}(0\ K;\,x)$ has smaller values and larger size variations than $\tau^e_{hh}(0\ K;\,x)$. The results are also consistent with the argument that the enhanced quantum confinement effect of the light holes leads to better carrier localization and recombination efficiency.

5. Conclusions

In summary, we have theoretically studied the quantum confinement effect on recombination dynamics and carrier localization in *c*-$In_xGa_{1-x}N$ low dimensional structures. Depending on the composition, temperature, and size of InN and $In_xGa_{1-x}N$ low dimensional structures, the quantum confinement effect can affect the exciton dimensions. In InN quantum cubes, the strong quantum confinement effect leads to temperature-dependent radiative lifetimes showing a large size variation. The nearly temperature-independent and shorter radiative lifetimes in small InN and In-rich $In_xGa_{1-x}N$ low dimensional structures suggest that the strong quantum confinement leads to *0* D carrier confinement, stronger carrier localization, and high recombination efficiency. Our simulation results indicate that spontaneous emissions come from the radiative recombination of localized excitons in In-rich $In_xGa_{1-x}N$ clusters. The simulation results could provide important information for the designs and interpretations of *c*-InN and *c*-$In_xGa_{1-x}N$ devices.

Acknowledgments

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