Radiative Recombination Lifetime and Exciton Dimension in Zinc Blende $Al_xGa_{1-x}N/AlN$ Quantum Boxes

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Received September 4, 2008; revised January 24, 2009; accepted February 2, 2009; published online May 20, 2009

In this work, to study the carrier dynamics and exciton dimensionality of the quantum confinement in zinc blende $Al_xGa_{1-x}N/AlN$ quantum boxes, oscillator strength and radiative recombination lifetime were theoretically calculated. In smaller quantum boxes, the feature of a transition region between zero dimension (0-D) and quasi-three dimension (3-D) at high temperatures implies that the strong quantum confinement effect leads to better carrier localization. Owing to the enhanced quantum confinement effect of light-hole eigenstates, transitions from electron to light-hole eigenstates were found to be more efficient than those from electron to heavy-hole eigenstates. In addition, because the effective masses of III–N semiconductors are higher than those of conventional III–V semiconductors, the stronger quantum confinement effect in smaller quantum boxes leads to larger band transition energy, shorter radiative lifetime, and better recombination efficiency. III–N semiconductors can be used to take advantage of the strong quantum confinement effect to develop low-dimensional nanostructures in optoelectronic devices.

DOI: 10.1143/JJAP.48.051001

1. Introduction

GaN-based low-dimensional (D) nanostructures have been extensively investigated in recent years because of their excellent optical and electrical properties.[1,2] Owing to their wide band gap, high thermal conductivity, high breakdown voltage, piezoelectric and pyroelectric properties, mechanical hardness, and chemical inertness, GaN-based compound semiconductors have become key materials in short-wavelength optoelectronics as well as the high-power electronics.[1,2] Most of the successive developments are due to the extensive utilization of low-D nanostructures in GaN-based devices. In low-D nanostructures, the movement of charge carriers is constrained by potential barriers. The excitons exhibit 0-D, 1-D, 2-D, or 3-D behavior depending on whether the potential barriers confine the carriers in quantum dots, quantum wires, quantum wells, or bulk structures, respectively.[3]

The temperature dependence of radiative lifetime can be used to test for the dimensionality of the quantum confinement in low-D nanostructures. For n-D excitons, radiative decay time is expected to depend on temperature as $(k_B T)^{n/2}$.[3,4] In zinc blende (ZB) GaN/AlN quantum dots (QDs), the temperature dependence of radiative lifetime shows dimensions between 0- and quasi-3-D.[5] Carrier thermalization between the ground and first excited states in such QDs was proposed to explain the dimension transition. In our previous studies, the quantum size effect was shown to be predominant in the optical properties of ZB $Al_xGa_{1-x}N/AlN$ and $In_xGa_{1-x}N/GaN$ quantum cubes.[6,7] Simulation results showed the importance of factors such as composition, size, quantum confined potential, effective mass, and strain in determining the size-dependent optical properties and electronic structures of such low-D nanostructures.[6,7] With the deconvolution of the quantum confined Stark effect in ZB GaN QDs, the quantum confinement and thermal effects on the carrier dynamics and exciton dimensions in size- and composition-dependent $Al_xGa_{1-x}N$ QDs can be fully understood.

In this work, to study the carrier dynamics and exciton dimensionality of the quantum confinement in ZB $Al_xGa_{1-x}N/AlN$ quantum boxes, radiative recombination lifetime was theoretically calculated. The temperature dependence of radiative recombination lifetime shows a hybrid of excition dimensions between 0- and quasi-3-D. Because the effective masses of III–N semiconductors are higher than those of conventional III–V semiconductors, the stronger quantum confinement effect in smaller quantum dots leads to larger band transition energies and shorter radiative lifetimes.

This paper is organized as follows: In §2, the theoretical calculations of the band structures and radiative recombination lifetimes of $Al_xGa_{1-x}N/AlN$ quantum boxes are described. In §3, we discuss the electronic structures of $Al_xGa_{1-x}N/AlN$ quantum boxes. In §4, oscillator strength, radiative recombination lifetime, and exciton dimension are discussed. Finally, conclusions are drawn in §5.

2. Theoretical Calculations

2.1 Band structures of $Al_xGa_{1-x}N/AlN$ quantum boxes

To calculate the band structure of a quantum box, we modified the discrete wave functions in the quantum box with an infinite barrier potential.[6–8] Composition- and size-dependent AlGaN quantum boxes, i.e., $L_x = L_y = L_z = L$, embedded in an AlN matrix were considered.[6,7] Electron and hole wavefunctions, $\psi_e(\boldsymbol{r}_e)$ and $\psi_h(\boldsymbol{r}_h)$, for the Schrödinger equations were considered, respectively:[6,7]

$$\left[-\frac{\hbar^2}{2}\nabla_e\cdot\left(\frac{1}{m_e^*(\boldsymbol{r}_e)}\right)\nabla_e + \Delta V_e(\boldsymbol{r}_e)\right]\psi_e(\boldsymbol{r}_e) = E_e\psi_e(\boldsymbol{r}_e), \quad (1a)$$

$$\left[-\frac{\hbar^2}{2}\nabla_h\cdot\left(\frac{1}{m_h^*(\boldsymbol{r}_h)}\right)\nabla_h + \Delta V_h(\boldsymbol{r}_h)\right]\psi_h(\boldsymbol{r}_h) = E_h\psi_h(\boldsymbol{r}_h). \quad (1b)$$

$\psi_e(\boldsymbol{r}_e)$ and $\psi_h(\boldsymbol{r}_h)$ can be expressed as the series expansions of the orthonormal functions $\phi_{l_e,m_e,n_e}(x,y,z)$ and $\phi_{l_h,m_h,n_h}(x,y,z)$ with the coefficients, d_{l_e,m_e,n_e} and d_{l_h,m_h,n_h}, respectively:

$$\psi_e(\boldsymbol{r}_e) = \sum_{l_e,m_e,n_e} d_{l_e,m_e,n_e}\phi_{l_e,m_e,n_e}(x,y,z), \quad (2a)$$

$$\psi_h(\boldsymbol{r}_h) = \sum_{l_h,m_h,n_h} d_{l_h,m_h,n_h}\phi_{l_h,m_h,n_h}(x,y,z), \quad (2b)$$

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Table I. Material parameters of GaN and AlN used for calculations.[9–12)]

	GaN	AlN
Lattice constant a (Å)	4.5	4.38
Band gap E_g^Γ (eV)	3.299	5.4
Effective mass (m_0)		
Electron effective mass	0.15	0.25
Heavy hole along c-axis	0.847	1.02
perpendicular to c-axis	1.526	2.439
Light hole along c-axis	0.236	0.349
perpendicular to c-axis	0.105	0.145
Momentum matrix element E_p (eV)	25	27.1
Hydrostatic deformation potential (eV)		
Conduction band a_c	−6.71	−4.5
Valence band a_v	−0.69	−4.9
Refractive index	2.6	2.1
Momentum matrix element (eV)	25	27.1

where $l_{\mathrm{e(h)}}$, $m_{\mathrm{e(h)}}$, and $n_{\mathrm{e(h)}}$ are positive integers denoting the quantum states in the conduction (valence) band. $\phi_{l_\mathrm{e},m_\mathrm{e},n_\mathrm{e}}(x,y,z)$ and $\phi_{l_\mathrm{h},m_\mathrm{h},n_\mathrm{h}}(x,y,z)$ are the eigenfunctions in the quantum box with infinite barrier potential. After substituting eqs. (2a) and (2b) into eqs. (1a) and (1b), we then multiplied both sides of eqs. (1a) and (1b) by $\phi_{l'_i,m'_i,n'_i}(x,y,z)$ (i = e, h) and integrated over the quantum box region (with extended dimensions). To extract the valence band mixing effect in the band structures, the averaged effective mass of the valence band was considered in the calculation.[9–11)] The hydrostatic strain between the quantum box and the outer material can modify the band structures. An eigenvalue problem of a $10 \times 10 \times 10$ tensor for either a conduction or valence band was solved. Details of the calculations were described elsewhere.[6,7)] The parameters of AlGaN except band-gap energy were calculated using the linear interpolation formula of AlN and GaN. Table I lists the physical parameters used for the calculations.[9–12)]

2.2 Radiative recombination lifetime

The radiative recombination lifetime and the dimensions of excitons in quantum boxes were analyzed. At 0 K, carriers relax to the ground state and only the ground state is considered. The radiative recombination lifetime $\tau(0\,\mathrm{K};x)$ for c-$\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{N}$ at 0 K can be expressed as[8)]

$$\tau(0\,\mathrm{K};x) = \frac{\pi\varepsilon_0\hbar^2c^3m_0^2(x)}{n(x)e^2E^{\mathrm{e}}_{\mathrm{hh(lh)}}(0\,\mathrm{K};x)|E_\mathrm{P}(x)|^2f(0\,\mathrm{K};x)} = \frac{\tau_\mathrm{c}(0\,\mathrm{K};x)}{f(0\,\mathrm{K};x)} \tag{3}$$

where $E^{\mathrm{e}}_{\mathrm{hh(lh)}}(0\,\mathrm{K};x)$ is the transition energy from the electron to heavy-hole (light-hole) eigenstates, $|E_\mathrm{P}(x)|^2$ 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. The composition-dependent $|E_\mathrm{P}(x)|^2$, $m_0(x)$, and $n(x)$ for c-$\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{N}$ were expressed as a linear interpolation formula of GaN and AlN.[8)] The oscillator strength, $f(0\,\mathrm{K};x)$, of the ground-state electron–hole wavefunction can be expressed as

$$f(0\,\mathrm{K};x) = \left|\sum_{\substack{l_\mathrm{e}=l_\mathrm{h}\\ m_\mathrm{e}=m_\mathrm{h}\\ n_\mathrm{e}=n_\mathrm{h}}} d^0_{l_\mathrm{e},m_\mathrm{e},n_\mathrm{e}}(0\,\mathrm{K};x)\,d^0_{l_\mathrm{h},m_\mathrm{h},n_\mathrm{h}}(0\,\mathrm{K};x)\right|^2. \tag{4}$$

It was assumed that the carrier density is lower than the box density. In III–N semiconductors, the carrier relaxation time is generally faster than the carrier recombination time.[13)] At higher temperatures, excitons are thermalized to higher excited levels. Boltzman distribution was used to average the oscillator strengths of the ground and higher excited states. The temperature-dependent oscillator strength, $f(T;x)$, is described as

$$f(T;x) = \frac{\sum_i f_i(0\,\mathrm{K};x)e^{-E_i(0\,\mathrm{K};x)/k_\mathrm{B}T}}{\sum_i e^{-E_i(0\,\mathrm{K};x)/k_\mathrm{B}T}}, \tag{5}$$

where $k_\mathrm{B}T$ is the thermal energy, E_i is the i-th eigenenergy, and $f_i(0\,\mathrm{K};x)$ is the i-th oscillator strength of the electron–hole wavefunction.[8)]

The temperature-dependent recombination lifetime is calculated as[8)]

$$\tau(T;x) = \frac{\tau_\mathrm{c}(0\,\mathrm{K};x)}{f(T;x)}. \tag{6}$$

For n-D excitons, the temperature-dependent oscillator strength depends on temperature as $(k_\mathrm{B}T)^{-n/2}$. Radiative recombination lifetime is inversely proportional to oscillator strength. Hence, for n-D excitons, radiative recombination lifetime is dependent on temperature as $(k_\mathrm{B}T)^{n/2}$.[3,4)]

3. Electronic Structures of $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{N}/\mathrm{AlN}$ Quantum Boxes

To investigate the quantum confinement effect on the electronic structures, the electron eigenstates ($|E^\mathrm{e}_n\rangle, n = 0,1,2,\ldots$), heavy-hole eigenstates ($|E^\mathrm{hh}_n\rangle, n = 0,1,2,\ldots$), and light-hole eigenstates ($|E^\mathrm{lh}_n\rangle, n = 0,1,2,\ldots$) were studied. Figures 1(a)–1(c) show the numbers of $|E^\mathrm{e}_n\rangle$, $|E^\mathrm{hh}_n\rangle$, and $|E^\mathrm{lh}_n\rangle$ as a function of aluminum composition, respectively. The number of eigenstates shows a large size and composition variations. Because of the enhanced quantum confinement effect in smaller quantum boxes, the number of eigenstates decreases. As the aluminum composition inside the boxes increases, the shallower quantum confinement potential confines fewer bound states. Hence, the number of eigenstates decreases. Among the three types of eigenstates, $|E^\mathrm{lh}_n\rangle$ has the smallest number of eigenstates and the strongest size dependence. This trend indicates that the quantum confinement effect is a critical factor in determining the electronic structures and the numbers of eigenstates in AlGaN quantum boxes.

4. Oscillator Strength, Radiative Recombination Lifetime, and Exciton Dimension

Figures 2(a)–2(c) shows the temperature dependence of the inverse of the oscillator strength, $1/f^\mathrm{e}_\mathrm{hh}(T;x)$, for GaN, $\mathrm{Al}_{0.4}\mathrm{Ga}_{0.6}\mathrm{N}$, and $\mathrm{Al}_{0.7}\mathrm{Ga}_{0.3}\mathrm{N}$ quantum boxes, respectively. The transitions between the electron and heavy-hole eigenstates are considered. The inverse of the oscillator strength provides much information on carrier dynamics. $1/f^\mathrm{e}_\mathrm{hh}(T;x)$ implies a hybrid of excition dimensions between 0- and quasi-3-D.[5,8,13)] In smaller boxes, $1/f^\mathrm{e}_\mathrm{hh}(T;x)$ is nearly

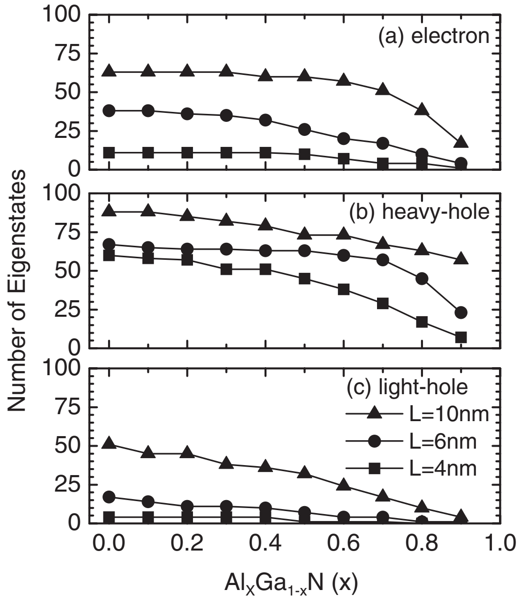

Fig. 1. Numbers of (a) electron, (b) heavy-hole, and (c) light-hole eigenstates as functions of aluminum composition for various box sizes.

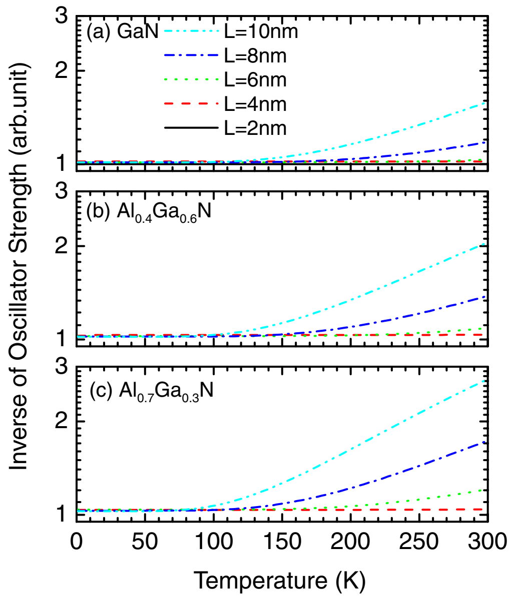

Fig. 3. (Color online) Temperature dependences of the inverse of oscillator strength, $1/f^{e}_{lh}(T;x)$, for (a) GaN, (b) $Al_{0.4}Ga_{0.6}N$, and (c) $Al_{0.7}Ga_{0.3}N$ quantum boxes. Transitions are between electron and light-hole eigenstates. $1/f^{e}_{lh}(T;x)$ is shown on the $\log_{10}$ scale for clarity.

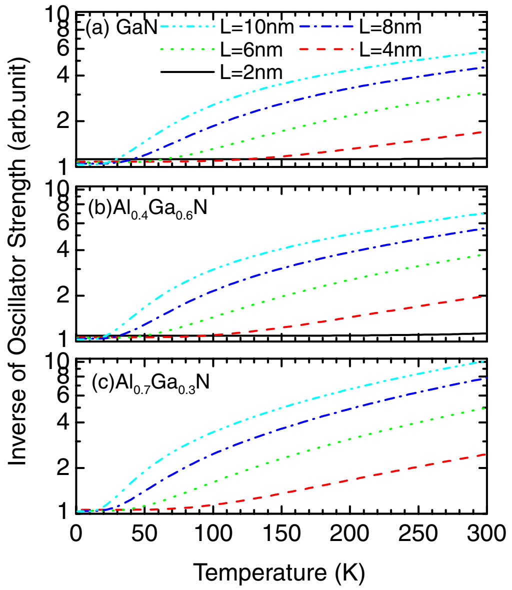

Fig. 2. (Color online) Temperature dependences of the inverse of oscillator strength, $1/f^{e}_{hh}(T;x)$, for (a) GaN, (b) $Al_{0.4}Ga_{0.6}N$, and (c) $Al_{0.7}Ga_{0.3}N$ quantum boxes. Transitions are between electron and heavy-hole eigenstates. $1/f^{e}_{hh}(T;x)$ is shown on the $\log_{10}$ scale for clarity.

constant and the transition region between 0- and quasi-3-D tends to shift to high temperatures. This implies a strong quantum confinement effect in smaller quantum boxes. Because of the relatively small number of discrete eigenstates in smaller boxes, the thermal distributions of excited states are not efficient. This leads to transition regions at high temperatures. In contrast, $1/f^{e}_{hh}(T;x)$ in large boxes shows larger temperature variations and transition regions at lower temperatures. Because of the larger number of discrete eigenstates in larger boxes, thermal excitation to the excited states is more efficient. The small oscillator strengths of excited states can account for the elongated $1/f^{e}_{hh}(T;x)$ at higher temperatures. In addition, $1/f^{e}_{hh}(T;x)$ decreases with increasing box size at low temperatures. This trend is similar to those of GaAs quantum boxes, ZB GaN/AlN QDs, and nonpolar GaN/AlN QDs, in which oscillator strength increases with increasing box lateral width and dot size.[5,8,14) At low temperatures, carriers relax to the ground state and only the ground state is considered. Owing to the strong quantum confinement effect in smaller quantum dots, the ground states shift upward and the wave function penetrates the AlN surrounding matrix. Because the small effective mass of electrons results in a stronger quantum confinement effect, the wave function penetration of electrons is larger than that of heavy holes. The smaller overlap integral of electron and hole wavefunctions reduces the oscillator strength. Hence, a smaller $1/f^{e}_{hh}(T;x)$ in larger quantum boxes at low temperatures is observed. Meanwhile, because thermal excitation to the excited states in larger boxes is more efficient, the small oscillator strengths of excited states can account for the elongated $1/f^{e}_{hh}(T;x)$ at higher temperatures. Furthermore, $1/f^{e}_{hh}(T;x)$ shows smaller size variations in GaN quantum boxes than in $Al_{0.4}Ga_{0.6}N$ and $Al_{0.7}Ga_{0.3}N$ quantum boxes. Both the strong quantum confinement effect and better quantum confined potential in GaN quantum boxes can explain the trends.

Figure 3 shows the temperature dependence of the inverse of the oscillator strength, $1/f^{e}_{lh}(T;x)$, for transitions between electron and light-hole eigenstates. Similar arguments to those for $1/f^{e}_{hh}(T;x)$ can explain the features of $1/f^{e}_{lh}(T;x)$. Owing to the light-hole eigenstates being more discrete, the transition region for $1/f^{e}_{lh}(T;x)$ between 0- and quasi-3-D shifts to higher temperatures. This suggests that the stronger quantum confinement effect of light-hole eigenstates leads to better carrier confinement. In addition, because fewer light-

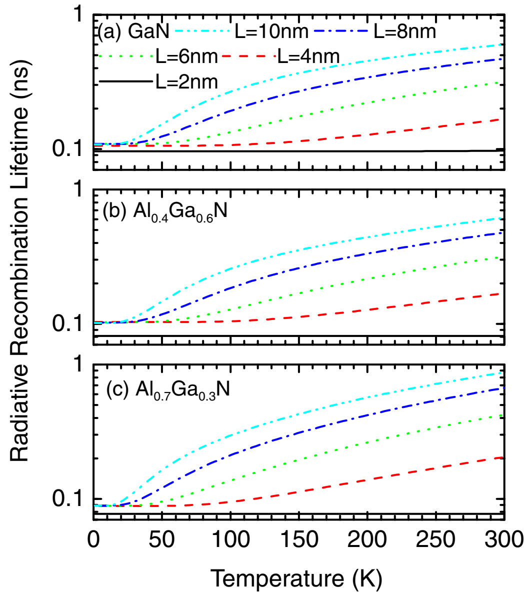

Fig. 4. (Color online) Temperature dependences of the radiative recombination lifetime, $\tau^{e}_{hh}(T;x)$, for (a) GaN, (b) $Al_{0.4}Ga_{0.6}N$, and (c) $Al_{0.7}Ga_{0.3}N$ quantum boxes. Transitions are between electron and heavy-hole eigenstates. $\tau^{e}_{hh}(T;x)$ is shown in $\log_{10}$ scale for clarity.

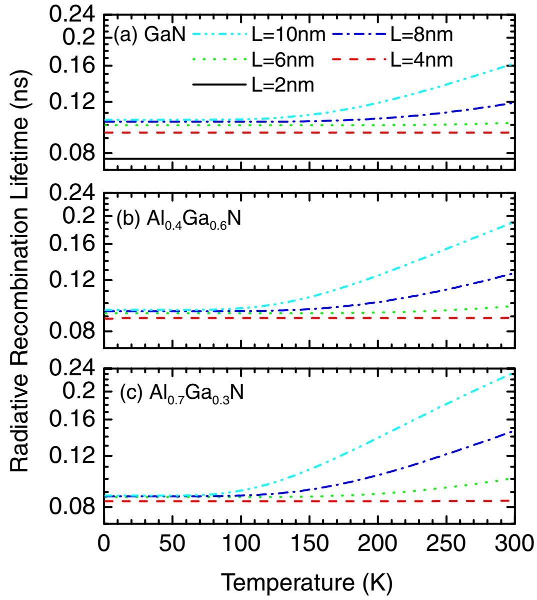

Fig. 5. (Color online) Temperature dependences of the radiative recombination lifetime, $\tau^{e}_{lh}(T;x)$, for (a) GaN, (b) $Al_{0.4}Ga_{0.6}N$, and (c) $Al_{0.7}Ga_{0.3}N$ quantum boxes. Transitions are between electron and light-hole eigenstates. $\tau^{e}_{hh}(T;x)$ is shown in $\log_{10}$ scale for clarity.

hole eigenstates result in the elongated inverse of oscillator strength, $1/f^{e}_{lh}(T;x)$ is smaller than $1/f^{e}_{hh}(T;x)$. It is noted that when the composition contrast or the box size is too small, the electron transitions do not occur.[6,7] Hence, electron transitions in $Al_{0.4}Ga_{0.6}N$ and $Al_{0.7}Ga_{0.3}N$ quantum boxes with 2 nm box size are absent.

Figures 4(a)–4(c) show the temperature dependences of the radiative recombination lifetime, $\tau^{e}_{hh}(T;x)$, for GaN, $Al_{0.4}Ga_{0.6}N$, and $Al_{0.7}Ga_{0.3}N$ quantum boxes, respectively. Transitions between electron and heavy-hole eigenstates are considered. $\tau^{e}_{hh}(T;x)$ consists of two regions. In low-temperature region, $\tau^{e}_{hh}(T;x)$ is a constant. Because only the ground state is occupied, the excitons exhibit 0-D behavior. In high-temperature region, $\tau^{e}_{hh}(T;x)$ is proportional to temperature as $T^{3/2}$. Carriers are thermally distributed to excited states and the quantum boxes exhibit quasi-3-D behavior. Depending on the composition, temperature, and size, the quantum boxes exhibit an intermediate spatial dimension between 0- and quasi-3-D. In small boxes, $\tau^{e}_{hh}(T;x)$ is nearly a constant and shows a transition region between 0- and quasi-3-D at high temperatures. This suggests that the strong quantum confinement in small boxes leads to a 0-D carrier confinement and a higher recombination efficiency. Furthermore, the smaller size variation and shorter $\tau^{e}_{hh}(T;x)$ in GaN quantum boxes imply better carrier confinement and recombination efficiency. In addition, our simulation results are in qualitative agreement with the trends of the experimental radiative lifetimes of ZB GaN/AlN QDs,[5] which indicates the reasonable assumption of the box shape in our simulation model.

Figure 5 shows the temperature-dependent radiative recombination lifetimes, $\tau^{e}_{lh}(T;x)$, for transitions from electron to light-hole eigenstates. Compared with $\tau^{e}_{hh}(T;x)$, $\tau^{e}_{lh}(T;x)$ shows a transition region between 0- and quasi-3-D at higher temperatures and shorter radiative recombination lifetimes. These features suggest that transitions from electron to light-hole eigenstates in quantum boxes show high recombination efficiency. By taking advantage of strain engineering to alter the valence band structure, the efficacy in radiative recombination can be improved.[15] In addition, because of the stronger quantum confinement effect in GaN quantum boxes than in $Al_xGa_{1-x}N$ quantum boxes, the following features of $\tau^{e}_{lh}(T;x)$ were observed: (1) a transition region between 0- and quasi-3-D at high temperatures, (2) nearly temperature-independent constants, and (3) small size variations.

Figure 6(a) shows the radiative lifetime, $\tau^{e}_{hh}(0\,K;x)$, of $Al_xGa_{1-x}N$ quantum boxes at 0 K for the transition energies, E^{e}_{hh}, from electron to heavy-hole eigenstates. It was shown that $\tau^{e}_{hh}(0\,K;x)$ decreases in smaller quantum boxes. This trend is consistent with the experimental radiative lifetimes of ZB and nonpolar GaN/AlN QDs.[5,14] Without the contribution of polarization fields in hexagonal GaN/AlN QDs, the radiative lifetimes of ZB GaN QDs are shorter than those in hexagonal GaN QDs. Our previous study shows that because of the strong quantum confinement effect, E^{e}_{hh} in smaller quantum boxes are larger than that in larger quantum boxes.[6,7] The larger E^{e}_{hh} in smaller quantum boxes leads to shorter radiative lifetimes. In conventional III–V semiconductors such as GaAs, the lifetime is longer in smaller quantum boxes.[8] Because of the larger effective mass and confinement potential depth, the quantum confinement effect in III–N semiconductors is stronger than that in GaAs semiconductors. In smaller quantum dots, the stronger quantum confinement effect leads to larger band transition energies and hence shorter radiative lifetimes were obtained. Therefore, III–N semiconductors can be used to take advantage of strong quantum confinement effect to develop low-D nanostructures in optoelectronic devices. In addition, although the smaller oscillator strengths in the smaller quantum boxes lead to a longer radiative lifetime, their values are similar and are close to unity. The contribution of smaller oscillator strengths to longer radiative lifetimes in

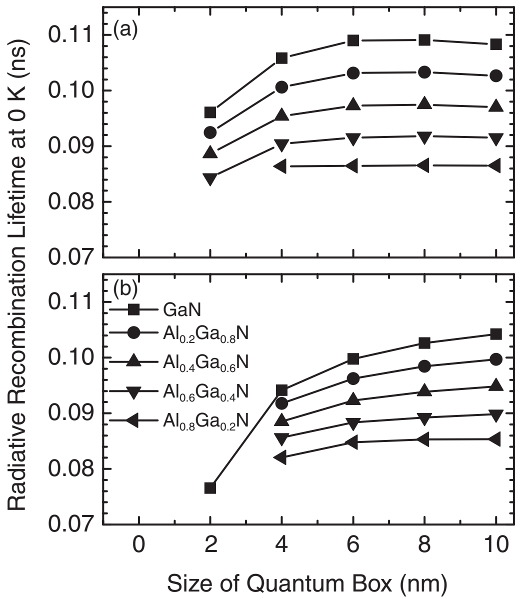

Fig. 6. (a) Radiative recombination lifetime, $\tau^{e}_{hh}(0\,K; x)$, as a function of box size at 0 K for transition from electron to heavy-hole eigenstates. (b) Radiative recombination lifetime, $\tau^{e}_{lh}(0\,K; x)$, as a function of box size at 0 K for transition from electron to light-hole eigenstates.

smaller quantum boxes is not apparent. Furthermore, owing to the strong quantum confinement effect and small band gap of GaN, E^{e}_{hh} displays pronounced variations with respect to the size of quantum cubes. From eq. (1), $\tau^{e}_{hh}(0\,K; x)$ of GaN quantum boxes shows larger size variations than that of AlGaN quantum boxes.

Figure 6(b) shows the radiative lifetime, $\tau^{e}_{lh}(0\,K; x)$, of $Al_xGa_{1-x}N$ quantum boxes at 0 K for the transition energy, E^{e}_{lh}, from electron to light-hole eigenstates. Owing to the enhanced quantum confinement effect of light-hole eigenstates, E^{e}_{lh} is larger than E^{e}_{hh} for the same box size and composition. The enhanced quantum confinement effect of light-hole eigenstates also leads to larger size variations of E^{e}_{lh}. From eq. (3), $\tau^{e}_{lh}(0\,K; x)$ is smaller values and has 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 light-hole eigenstates leads to better carrier localization and recombination efficiency.

5. Conclusions

In summary, the recombination dynamics and exciton dimensionality in $Al_xGa_{1-x}N/AlN$ quantum boxes were studied. The quantum boxes exhibit an intermediate spatial dimension between 0- and quasi-3-D. Because the effective masses of III–N semiconductors are higher than those of conventional III–V semiconductors, the stronger quantum confinement effect in smaller quantum boxes leads to larger band transition energies and shorter radiative lifetimes. III–N semiconductors can be used to take advantage of the strong quantum confinement effect to develop low-D nanostructures in optoelectronic devices. In addition, the simulation results are generally in agreement with the reported experimental results of cubic GaN/AlN quantum dots, which indicates the reasonable assumption of the cube shape in our simulation model.

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