Radiative life time of an exciton confined in a strained $GaN/Ga_{1-x}Al_xN$ cylindrical dot: built-in electric field effects*

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The binding energy of an exciton in a wurtzite GaN/GaAlN strained cylindrical quantum dot is investigated theoretically. The strong built-in electric field due to the spontaneous and piezoelectric polarizations of a GaN/GaAlN quantum dot is included. Numerical calculations are performed using a variational procedure within the single band effective mass approximation. Valence-band anisotropy is included in our theoretical model by using different hole masses in different spatial directions. The exciton oscillator strength and the exciton lifetime for radiative recombination each as a function of dot radius have been computed. The result elucidates that the strong built-in electric field influences the oscillator strength and the recombination life time of the exciton. It is observed that the ground state exciton binding energy and the interband emission energy increase when the cylindrical quantum dot height or radius is decreased, and that the exciton binding energy, the oscillator strength and the radiative lifetime each as a function of structural parameters (height and radius) sensitively depend on the strong built-in electric field. The obtained results are useful for the design of some opto-photoelectronic devices.

Keywords: quantum dot, donor bound excitons, oscillator strength

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1. Introduction

Among the wide band gap semiconducting materials, GaN/AlN semiconductors are promising heterostructures for variable wavelength devices due to their high luminescence efficiency[1,2] and can be realized for future spin-based spintronic devices. GaN-based diluted magnetic semiconductors are promising candidates for spin injection because of their expected good matching to AlGaN/GaN heterostructures.[3] Even at room temperature, ferromagnetism can be achieved in high band-gap semiconductors such as GaN and ZnO. So there has been recent interest in exploring AlGaN/GaN heterostructures for spintronic applications.[4] It is an ideal material for room temperature light emitting diodes and laser diodes due to the large difference in band gap energies between these two materials. Furthermore, these materials possess a high built-in electric field, of the order of MV/cm, induced by polarization in the (0001) growth direction[5] due to polarization discontinuity along the *c*-axis at the well barrier interface. This built-in internal electric field is very sensitive to the exact geometry of the system such as shape and strain. Moreover, these materials may have large lattice-mismatch strain fields, leading to the presence of large piezoelectric fields and a large density of dislocations, which may affect the optical properties. The optical band gap, their composition with strain dependence and their large difference between the absorption and the emission spectrum will lead to a drastic change in optical properties. Thus, the strain of the heterostructures and the density of the defects will exert a great influence on their physical properties. Therefore, GaN-based quantum heterostructures have attracted much attention due to their potential device applications.

Wurtzite GaN quantum dots (QDs) have been extensively studied due to their potential technological applications in optical, optoelectronic and electronic devices.[6] The optical and structural properties of doped GaN QDs have been reported earlier.[7] Many experimental and theoretical studies[8–10] have

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been undertaken regarding the wurtzite GaN QD due to the existence of its strong built-in internal electric field, which eventually leads to a reduction in the effective band gap of GaN QDs and affects the exciton state and luminescent properties.[11]

In the present paper, we concentrate on the behaviours of exciton binding energy and the radiative lifetime of an exciton of a strained GaN/$Ga_{1-x}Al_xN$ cylindrical QD with and without the effect of built-in electric field. The built-in electric field due to piezoelectricity and spontaneous polarization of a GaN/AlN QD and the anisotropy of the wurtzite crystal structures and the strains of the QD are included. They have been computed, in the single-band effective mass approximation, by changing the structural parameters and taking into account the valence-band anisotropy through the energy dependent mass in different spatial directions. The rest of the present paper is organized as follows. In Section 2, we outline the theory of the QD model and exciton binding energy. In Section 3, we discuss our numerical results. Finally, the main conclusions obtained in this paper are summarized in Section 4.

2. Theory and model

In this problem we assume that the GaN QD is an isolated cylindrical shape with radius R and height H along the z direction, surrounded by a large energy gap material, GaAlN. In the effective mass-approximation, the Hamiltonian of an exciton in a GaN/GaAlN QD, consisting of a single electron part ($H_{\rm e}$), the single hole part ($H_{\rm h}$) and Coulomb interaction term between electron–hole pair, is given by

$$H_{\rm ex} = H_{\rm e} + H_{\rm h} - \frac{e^2}{\varepsilon\,|r|}, \tag{1}$$

where e is the absolute value of a electron charge, $|\bar{r}| = |\bar{r}_{\rm e} - \bar{r}_{\rm h}|$ denotes the relative distance between the electron and the hole, and $\varepsilon = 10.4$ is the GaN dielectric constant of material inside the QD.

The Hamiltonian of the electron (hole) in the cylindrical co-ordinates is given by

$$H_j = -\frac{\hbar^2}{2m_j^*}\left[\frac{\partial^2}{\partial\rho_j^2} + \frac{1}{\rho_j}\frac{\partial}{\partial\rho_j} + \frac{1}{\rho_j^2}\frac{\partial^2}{\partial\varphi^2} + \frac{\partial^2}{\partial z_j^2}\right] + V(\rho_j, z_i) \mp eFz_j, \tag{2}$$

where the subscript j = e, h refers to the electron or hole, F is the built-in electric field, and the sign $\mp$ is for the electron (hole).

The effective mass m_j^* of the electron (hole) is given by

$$m_j^* = \begin{cases} m_{{\rm I}j}^*, & \rho_j \le R, \\ m_{{\rm II}j}^*, & \text{otherwise}, \end{cases} \tag{3}$$

where $m_{{\rm I}j}^*$ denotes the effective mass of GaN, and $m_{{\rm II}j}^*$ denotes the effective mass of the outer material, GaAlN. The material-dependent effective mass has been used in this calculation since it has a large influence on the electron (hole) energy level in semiconductor QDs, especially for narrow dots.[12] The electron (hole) confinement potential $V(\rho_j, z_j)$ due to the band offset in the GaN/ GaAlN QD structure is given by

$$V(\rho_j, z_j) = \begin{cases} V(\rho_j), & -H/2 \le z \le H/2, \\ V_{{\rm II}j}, & \text{otherwise}, \end{cases} \tag{4}$$

$$V(\rho_j) = \begin{cases} 0, & \rho_j \le R, \\ V_{{\rm I}j}, & \text{otherwise}, \end{cases} \tag{5}$$

where H is the height of the cylindrical QD. We have taken the band gap as[13]

$$E_{{\rm Ga}_{1-x}{\rm Al}_x{\rm N}}(x) = 6.2x + 3.4(1-x) + x(1-x){\rm eV}. \tag{6}$$

The material parameters are given in Table 1, and the band offset is taken to be 75:25. The energy-dependent effective mass is given by[14]

$$\frac{1}{m(E)} = \frac{1}{m_0}\frac{E_{\rm g}(E_{\rm g}+\Delta)}{(3E_{\rm g}+2\Delta)}\left[\frac{2}{E+E_{\rm g}} + \frac{2}{E+E_{\rm g}+\Delta}\right], \tag{7}$$

where ε is the dielectric constant of GaN, E denotes the electron energy in the conduction band, m_0 is the free electron mass, $E_{\rm g}$ and Δ are the band gap energy of GaN and spin-orbit band splitting, respectively. In addition,

$$g(E) = 2\left[1 - \frac{m_0}{m(E)}\frac{\Delta}{3(E_{\rm g}+E)+2\Delta}\right] \tag{8}$$

is the effective Lande factor of the electron (hole).[15]

Table 1. The parameters used in the calculation.

Parameter	GaN	$Ga_{1-x}Al_xN$
m^*/m_0	0.2	$0.3x + 0.2(1-x)$
$E_{\rm g}$/eV	3.4	$3.4(1-x) + 6.2x + 1.0x(1-x)$
a/nm	0.3189	$0.3189(1-x) + 0.311x$
e_{31}/C·m^{-2}	–0.2	0.48
e_{33}/C·m^{-2}	–0.29	1.59
$C_{13}/10^{10}$ N·m^{-2}	11.4	—
$C_{33}/10^{10}$ N·m^2	38.1	—
γ_1	2.67	1.92
γ_2	0.75	0.47
ε	10.6	8.31

The strain effects will induce an extra potential field, $V_{\rm strain}$. For the strained QD nanostructure, the confinement potential can be written as a sum of energy offsets of the conduction band (or valence band), $V_{\rm band}(\rho_j, z_j)$, and the strain-induced potential, $V_{\rm strain}(\rho_j, z_j)$, as

$$V(\rho_j, z_j) = V_{\rm band}(\rho_j, z_j) + V_{\rm strain}(\rho_j, z_j). \quad (9)$$

The electron (hole) confinement potential $V(\rho_j, z_i)$ due to the band offset in the GaN/GaAlN QD structure is given by

$$V_{\rm band}(\rho_j, z_j) = \begin{cases} 0, & \rho_j \le R, \\ V_j^0, & \text{otherwise}, \end{cases} \quad (10)$$

where V^0 is the barrier binding the carrier in the wire, which is taken as $V_{\rm e}^0 = 2828$ meV and $V_{\rm h}^0 = 942$ meV for $x = 0.1$ in $Ga_{1-x}Al_xN$.

The lattice mismatch between the GaN dot and the GaAlN barrier material induces a biaxial strain in the z- plane, lifting the degeneracy of the valence band. The strain-induced potential for the conduction band can be expressed as[16]

$$V_{\rm strian} = a_{\rm c}(\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}). \quad (11)$$

Here, $a_{\rm c}$ is the deformation potential constant of the conduction band, $\varepsilon_{xx} = \varepsilon_{yy} = (a_0 - a)/a$, where a_0 and a are the lattice parameters of bulk GaN and GaAlN respectively, and $\varepsilon_{zz} = -2(C_{12}/C_{11})\varepsilon_{xx}$, the values of parameters are given in Table 1.

The strain-induced potential for the valence band can then be written as[17]

$$V_{\rm strain} = a_{\rm v}(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz}) - \frac{b}{2}(\varepsilon_{xx}+\varepsilon_{yy}-2\varepsilon_{zz}), \quad (12)$$

where $a_{\rm v}$ and b are the deformation potential constants of valence band.

The polarization in a self-formed wurzite QD will have both a spontaneous component and a piezoelectric component. The piezoelectric tensor will have three independent nonvanishing components (e_{31}, e_{33}, e_{15}) but the z component of the spontaneous polarization is nonzero.[18] Thus, the total polarization, $\boldsymbol{P}_{\rm tot}$, is the sum of the spontaneous polarization $\boldsymbol{P}_{\rm SP}$ and the piezoelectric polarization $\boldsymbol{P}_{\rm PE}$ without the external electric field. The built-in electric field is caused by the polarization of the QD. Then from Ref. [19], we have

$$F_{\rm GaAlN} z_{\rm GaAlN} + F_{\rm GaN} z_{\rm GaN} = 0. \quad (13)$$

Using the boundary condition of the continuity of the electric displacement vector at the heterointerfaces of our QD, one can obtain

$$\varepsilon_{\rm e(GaN)}\varepsilon_0 F_{\rm GaN} - \varepsilon_{\rm e(GaAlN)}\varepsilon_0 F_{\rm GaAlN} = P_{\rm SP(GaAlN)} + P_{\rm PE(GaAlN)} - P_{\rm SP(GaN)}, \quad (14)$$

where ε_0 is the dielectric constant of the respective material. Using Eqs. (13) and (14), we can obtain the built-in electric field as

$$F_{\rm GaN} = \frac{(P_{\rm SP(GaAlN)} + P_{\rm PE(GaAlN)} - P_{\rm SP(GaN)}) z_{\rm GaAlN}}{\varepsilon_0(\varepsilon_{\rm e(GaN)} z_{\rm (GaAlN)} + \varepsilon_{\rm e(GaAlN)} z_{\rm (GaN)})}, \quad (15)$$

and

$$F_{\rm GaAlN} = -\frac{(P_{\rm SP(GaAlN)} + P_{\rm PE(GaAlN)} - P_{\rm SP(GaN)}) z_{\rm GaN}}{\varepsilon_0(\varepsilon_{\rm e(GaN)} z_{\rm (GaAlN)} + \varepsilon_{\rm e(GaAlN)} z_{\rm (GaN)})}. \quad (16)$$

The direction of the built-in electric field F depends on the orientation of the piezoelectricity and spontaneous polarization. In this case, the directions of the piezoelectricity and spontaneous polarization are both along the z direction.

The wave function of the electron (hole) confined in a GaN /GaAlN QD can be written as

$$\psi_j(\rho_j, \varphi_j, z_j) = f(\rho_j) h(z_j) \, {\rm e}^{{\rm i}m\varphi_j}, \quad j = 0, \pm 1, \pm 2, ..., \quad (17)$$

where m is the electron (hole) z-component angular momentum quantum number. The radial wave function $f(\rho_j)$ and the corresponding confinement energy equation of the electron (hole) can be obtained using the m-order Bessel function $\rm J_m$ and the modified Bessel function $\rm K_m$. The z-axis wave function $h(z_j)$ can be found using the linear combination of analytical functions $\sin(\xi)$ and $\cos(\xi)$ (inside the dot), or $\exp(\xi)$ (outside the dot).

The anisotropy and the corrections due to the conduction band non-parabolicity are important since the parallel mass determines the electron energy in the xy plane and the perpendicular mass determines the quantization energy of the electron in the z-plane. In the valence band of semiconductors, there are heavy hole and light hole bands, which degenerate on the top of the valence band. The valence-band anisotropy is described by assuming different hole masses in different directions. The masses in each direction of the same corresponding to the curvature of the hole bands around the Γ point, i.e., for the heavy-hole and light-hole are respectively[20]

$$\frac{m_0}{m_{\rm hh}^*} = \gamma_1 - 2\gamma_2, \quad (18)$$

$$\frac{m_0}{m^*_{\rm lh}} = \gamma_1 + 2\gamma_2, \tag{19}$$

where γ_1 and γ_2 are Luttinger parameters (see Table 1).

Since the inclusion of an impurity potential leads to a nonseparable differential equation, which cannot be solved analytically, it is necessary to use a variational approach to calculate the eigenfunction and eigenvalue of the Hamiltonian to calculate the bound exciton ground state energy. Considering the correlation of the electron–hole relative motion, we can choose the trial wave function as

$$\Psi(\bar{r}_{\rm e}, \bar{r}_{\rm h}) = \psi_{\rm e}(\rho_{\rm e}, \varphi_{\rm e}, z_{\rm e})\psi_{\rm h}(\rho_{\rm h}, \varphi_{\rm h}, z_{\rm h})\, {\rm e}^{-\alpha\rho^2}\, {\rm e}^{-\beta z^2}, \tag{20}$$

where $\psi_{\rm e}$ and $\psi_{\rm h}$ are the electron and hole wave functions, respectively, as explained earlier for the QD. The above equation describes the correlation of the electron–hole relative motion. α and β are the variational parameters responsible for the in-plane correlation and the correlation of the relative motion in the z-direction, respectively.[21] It should be pointed out that the two-parameter variational wave function is a reasonable choice for studying the impurity states in wurtzite GaN/GaAlN a QD. Therefore, wave function Eq. (16) completely describes the correlation of the electron–hole relative motion.

The ground state energy of the exciton in the GaN/GaAlN QD is calculated using the following equation:

$$E_{\rm ex} = \min_{\alpha,\beta} \frac{\langle \psi_{\rm ex} | H_{\rm ex} | \psi_{\rm ex} \rangle}{\langle \psi_{\rm ex} | \psi_{\rm ex} \rangle}. \tag{21}$$

The donor exciton binding energy $E_{\rm b}$ and the interband emission energy $E_{\rm ph}$ associated with the exciton are calculated using the following equations:

$$E_{\rm b} = E_{\rm e} + E_{\rm h} - E_{\rm ex}, \tag{22}$$

$$E_{\rm ph} = E_{\rm e} + E_{\rm h} + E_{\rm g} - E_{\rm ex}, \tag{23}$$

where $E_{\rm e}$ and $E_{\rm h}$ are the confinement energies of the electron and the hole, respectively.

3. Results and discussion

Numerical calculations have been carried out to calculate the donor exciton binding energy, the oscillator strength and its radiative lifetime in a cylindrical GaN/GaAlN strained dot with a heavy hole mass, because heavy excitons are more common in experimental results. The exciton binding energy values are computed as a function of dot radius for different heights of the cylindrical dot. We have also investigated the effect of the built-in electric field on the charged exciton when the dot radius is changed. The material parameters used in this study are listed in Table 1 and taken from Ref. [11]. The units of length and energy used throughout the present paper are the effective Bohr radius $R^* = \hbar^2\varepsilon/m^*e^2$ and the effective Rydberg $R^*_y = m^*e^4/2\varepsilon^2\hbar^2$, where m^* is the effective mass of the electron in the conduction band minimum of GaN.

Figure 1 shows the variations of exciton binding energy as a function of dot radius of a GaN/GaAlN cylindrical QD for two different dot heights (50 Å and 30 Å) without the inclusion of a built-in electric field. In all the cases, we have observed that the binding energy increases with the decrease of dot radius, reaching a maximum value and then decreasing when the dot radius further decreases. An increase in the dot radius results in a spreading of the wave function and so causes the lowering of the binding energy. As the radius of dot approaches zero, the confinement becomes negligibly small and in the finite barrier problem the tunneling becomes huge. This results in the decrease in binding energy as the dot radius becomes small. In addition, we observe that the exciton binding energy is reduced when the height of the QD increases. This is due to the increase of the relative distance between the electron and hole, which leads to the decrease of the Coulomb interaction with the increase of height H. This effect is in good agreement with previous investigations.[22]

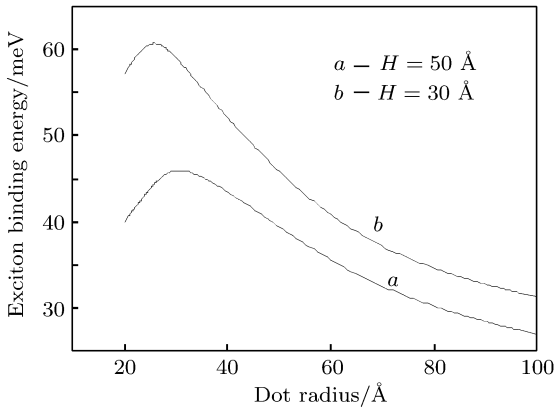


Fig. 1. Variations of exciton binding energy as a function of dot radius of a GaN/GaAlN cylindrical QD for two different dot heights.

We display the variations of exciton binding energy as a function of dot radius of a GaN/GaAlN cylindrical QD of height 50 Å with and without the effect of a built-in electric field in Fig. 2. The electric field ef-

fect on the ground-state donor binding energy is investigated. We observe that the exciton binding energy is reduced with the inclusion of the built-in electric field. This is because the electric field pushes the electron and the hole to the opposite sides, reducing their overlap and the exciton binding energy. Moreover, we notice that this effect is more pronounced for narrow dots due to the confinement. The difference in binding energy for a 30 Å GaN QD with and without the built-in electric field is over 10 meV. Hence, the built-in electric field has a large influence on the exciton states confined in the GaN QDs.

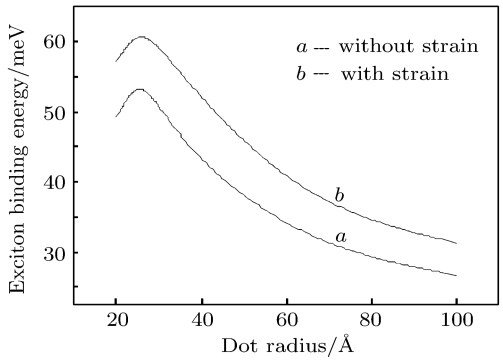


Fig. 2. Variations of exciton binding energy as a function of dot radius of a GaN/GaAlN cylindrical QD of height 50 Å with and without the effect of a built-in electric field.

Figure 3 shows the variations of interband emission energy as a function of dot radius of a GaN/GaAlN cylindrical QD of height 50 Å, with and without the strain effect. This representation clearly brings out the quantum size effect. The interband emission energy is decreased monotonically as the radius of the dot is increased for the GaN/GaAlN cylindrical QD. This is due to the confinement of electron–hole with respect to the z-plane when the dot radius is increased. Furthermore, we notice that the effect of the built-in electric field reduces the interband emission energy for all the dot radii due to the reduction in exciton binding energy.

Figure 4 shows the variations of oscillator strength of the exciton as a function of the dot radius of a GaN/GaAlN cylindrical QD of height 50 Å with and without the effect of a built-in electric field. In absorption spectra, the intensity of an exciton binding energy and the envelope wave function are characterized by the oscillator strength[23]

$$f = \frac{E_{\rm p}}{E_{\rm exc}} \left| \int_V \psi_{\rm exc}(r)\,{\rm d}r \right|^2, \tag{24}$$

where the Kane energy of GaN is $E_{\rm p} = 14$ meV.[24] It is observed that the oscillator strength of the exciton decreases as the dot radius increases in all cases. The oscillator strength of the exciton increases as the dot radius increases in both cases. This is due to the fact that the recombination rate of the electron and hole increases slowly with the increase of dot radius. Moreover, it is observed that the effect of the built-in electric field increases the oscillator strength. We have obtained the oscillator strength of the exciton $f = 28$ for a 20 Å GaN/GaAlN QD, which is in good agreement with another result.[25]

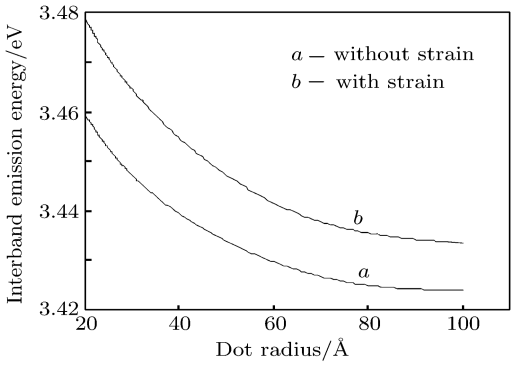


Fig. 3. Variations of interband emission energy as a function of dot radius of a GaN/GaAlN cylindrical QD of height 50 Å, with and without the effect of a built-in electric field.

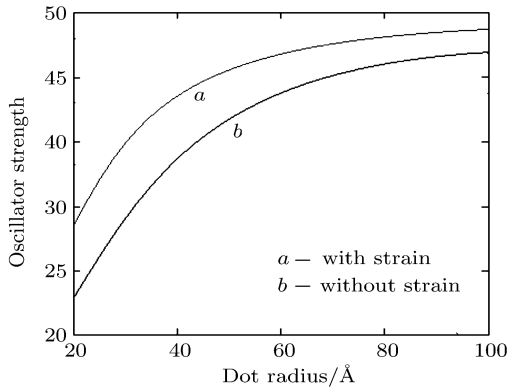


Fig. 4. Variations of oscillator strength as a function of dot radius of a GaN/GaAlN cylindrical QD of height 50 Å with and without the effect of a built-in electric field.

Figure 5 displays the variations of radiative life time as a function of dot radius of a GaN/GaAlN cylindrical QD of height 50 Å, with and without the effect of a built-in electric field. The radiative life time can be calculated as[26]

$$\tau = \frac{2\pi\varepsilon_0 m_0 c^3 h^2}{n e^2 E_{\rm exc}^2 f}, \tag{25}$$

where $E_{\rm exc}$ is the exciton binding energy, and all the other parameters are universal physical constants

and n is the refractive index which we have taken to be 2.29. It is found that the radiative lifetime of the exciton increases with the dot radius in all cases.

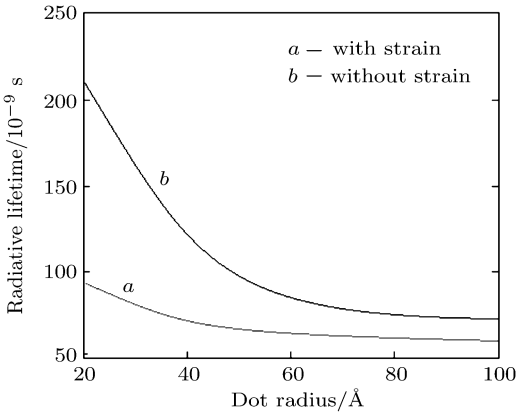


Fig. 5. Variations of radiative life time as a function of dot radius of a GaN/GaAlN cylindrical QD of height 50 Å, with and without strain effect.

The built-in electric field reduces the radiative lifetime for all dot radii. For a 20 Å GaN/GaAlN QD, we have observed the radiative lifetime to be 90 ps with the inclusion of an internal electric field and it is recorded to be 210 ps in the absence of an internal electric field. Simon *et al.*[27] have measured the lifetime to be 200 ps in cubic phase GaN/AlN QD without an internal electric field.

4. Conclusion

Within the framework of the effective-mass approximation, exciton states confined in wurtzite GaN/GaAlN strained QDs are investigated by a variational approach, including the strong built-in electric field effects caused by piezoelectricity and spontaneous polarization. The exciton binding energies have been computed by changing the structural parameters, taking into account the valence-band anisotropy through energy dependent mass in different spatial directions. Numerical results show that the exciton binding energies and the interband emission energy are both increased when the height or radius of the cylindrical QD is decreased. We hope that our investigations will play an important role in determining opto-electronic devices based on GaN/GaAlN QD nanostructures in the near future.

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