Excitonic Properties of GaN/AlN Quantum Dot Single Photon Sources

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***Abstract*—Excitons and biexcitons in GaN/AlN quantum dots (QD) were investigated with special emphasis on the use of these QDs for single photon source applications. The theoretical methodology for the calculation of single-particle states was based on 8-band strain-dependent envelope function Hamiltonian, with the effects of spin-orbit interaction, crystal-field splitting, piezoelectric and spontaneous polarization taken into account. Exciton and biexciton states were found using the configuration interaction method. Optimal QD heights for their use in single-photon emitters were determined for various diameter to height ratios. The competition between strong confinement in GaN QDs and internal electric field, generally reported in wurtzite GaN, was also discussed, as well as its effect on appearance of bound biexcitons.**

I. Introduction

Modern optoelectronic devices like triggered single-photon sources ("photon on demand") are highly desired for applications in quantum cryptography and quantum information processing [1]. GaN/AlN quantum dots [2], [3] offer certain advantages for realization of single photon sources. Larger band offsets and effective masses lead to strong quantum confinement effects, which should enable the operation of single photon sources (SPS) at higher temperatures. Several single III-nitride quantum dot spectroscopy experiments were therefore performed, [4], [5], [6] which indeed led to the realization of a GaN/AlN single photon source operating at 200 K. [7] For SPS applications it is desirable to have as large as possible the value of biexcitonic shift defined as the difference between the energy of the transition line from the biexciton to exciton state and the energy of the exciton transition line, $B_{\rm XX} = E_{\rm XX} - 2E_{\rm X}$. This is required to enable good spectral separation of the two lines. [7] It is known [3] that the built-in electric field acts to localize the electrons at the top of the dot and holes at the bottom of the dot. Consequently, the interaction between two excitons forming a biexciton is mostly determined by repulsive electron-electron and hole-hole interactions which are stronger than the attractive interaction between spatially separated electron and hole. [6] For quantum dots with larger heights the biexciton is therefore certainly unbound and biexcitonic shift increases as the height increases due to a decreasing attractive part of the interaction. From that perspective, it is desirable to have a large QD height. On the other hand, one should also have the optical transition matrix element of the exciton transition as large as possible. [8] For large QDs, this element is small due to spatial separation of electron and hole wavefunctions and it is therefore desirable to have a small QD height from this perspective. This discussion therefore indicates that the appropriate QD geometry for single photon source applications should be determined as a compromise between the two opposite requirements, which requires detailed knowledge of the excitonic properties of these dots.

II. Theoretical method

After the single particle states were found, using the multiband **k·p** Hamiltonian, the (bi)exciton states were obtained using the configuration interaction (CI) method. [9], i.e. by direct diagonalization of the Hamiltonian

$$\begin{aligned}\hat{H} &= \sum \varepsilon_i \hat{e}_i^+ \hat{e}_i - \sum \varepsilon_i \hat{h}_i^+ \hat{h}_i + \frac{1}{2}\sum V_{iljk} \hat{e}_i^+ \hat{e}_j^+ \hat{e}_k \hat{e}_l \\ &+ \frac{1}{2}\sum V_{iljk} \hat{h}_i^+ \hat{h}_j^+ \hat{h}_k \hat{h}_l - \sum (V_{iljk} - V_{ikjl}) \hat{e}_i^+ \hat{h}_j^+ \hat{h}_k \hat{e}_l \end{aligned} \quad (1)$$

where $\hat{e}$ ($\hat{e}^+$) are electron annihilation (creation) operators, $\hat{h}$ ($\hat{h}^+$) the same operators for holes, and ε_i the single-particle energies. The summation over each index takes place over electron or hole states only, depending whether that index corresponds to electron or hole operator. Coulomb integrals, V_{ijkl}, required for the diagonalization of the CI Hamiltonian were evaluated in reciprocal space and then corrected using the Makov-Payne method [10], [9] by adding the first few terms (monopole, dipole, and quadrupole) in the multipole expansion to compensate for the effect of the mirror charges induced by periodic boundary conditions. These read as

$$\begin{aligned} V_{ijkl} &= V_{ijkl}(\Omega_c) - \frac{e^2}{4\pi\varepsilon}[q_{ij}q_{kl}a_{\rm mad} + \frac{4\pi}{3\Omega_c}\boldsymbol{d}_{ij}\cdot\boldsymbol{d}_{kl} \\ &- \frac{2\pi}{3\Omega_c}(q_{ij}Q_{kl} + q_{kl}Q_{ij})], \end{aligned} \quad (2)$$

where $V_{ijkl}(\Omega_c)$ is uncorrected Coulomb integral calculated on supercell Ω_c, and $q_{ij}(\Omega_c) = \delta_{ij}$, $\boldsymbol{d}_{ij}(\Omega_c)$, and $Q_{ij}(\Omega_c)$ are the monopole, dipole and quadrupole corrections respectively, that acquires analytic form in the PW representation. The Madelung term in Eq. (2), $a_{\rm mad}$, is defined via Ewald sums in real and inverse space, and self-interaction correction. Depending on order of indices in Eq. (2) those integrals represent direct Coulomb integrals $J_{ab} = V_{aabb}$ or exchange Coulomb integrals $K_{ab} = V_{abab}$. An efficient and accurate method to evaluated these expressions in reciprocal space was described in Ref. [9]. Additionally, symmetry considerations imply that only Coulomb integrals V_{ijkl} whose wavefunctions satisfy the conservation of the total quasi-angular momentum,

$\{m_j+m_l \equiv m_i+m_k (\mathrm{mod} 6)\}$ are nonzero. These are therefore the only ones that need to be evaluated, which reduces the number of integrals that need to be calculated by a factor of six. The whole methodology presented here was implemented in the kppw code. [11]

III. Results and Discussion

The calculations of the single particle electron and hole states and of the excitonic structure have been performed for a set of QDs satisfying the following conditions. The QD height was varied in the range $h = 1.5\mathrm{nm}$ to $h = 5\mathrm{nm}$ with a step of $0.5\mathrm{nm}$. The diameter to height ratio D/h was varied from 4 to 10 with a step of 1, and dots with the diameter larger than 30 nm were discarded. The truncated hexagonal pyramid base angle of $\alpha = 30^{\mathrm{o}}$ was assumed and the wetting layer width of 0.5185 nm. All CI calculations used the basis set consisting of $N_e = 8$ electron and $N_h = 14$ hole states. For all D/h ratios we get an expected result that the exciton energy E_{X} and biexcitons E_{XX} decrease as the quantum dot height is increased [12]. We have observed that for maximizing the biexciton shift, B_{XX}, smaller values of D/h ratio are required, also in agreement with the experimental findings of Ref. [7]. Within the fixed value of D/h, bigger dots tend to have larger values of the B_{XX} due to reduction of the attractive part of the Coulomb interaction. Unfortunately, the trends in dipole matrix elements, required for the bright emission from the SPS, are opposite to the trends in bi-exciton shifts. Therefore, a compromise between these trends has to be made to find the optimal quantum dot geometry. To achieve this, we define the optimization function as [13], [12]:

$$\Xi = (E_{\mathrm{XX}} - 2E_{\mathrm{X}}) \cdot \ln(p_{\mathrm{X}}^{(x)}/p_{\mathrm{X}}^{(0)}). \qquad (3)$$

$p_{\mathrm{X}}^{(x)}$ is the value of the $x-$component of the dipole matrix element of the exciton transition, $p_{\mathrm{X}}^{(0)}$ is equal to $10^{-4}p_{\mathrm{X}}^{(x),\mathrm{max}}$ and $p_{\mathrm{X}}^{(x),\mathrm{max}}$ is the maximal value of $p_{\mathrm{X}}^{(x)}$ for all quantum dots considered. While the choice of $p_{\mathrm{X}}^{(0)}$ is somewhat arbitrary, we find that the positions of maxima of the optimization function are weakly dependent on its value, when it is changed within reasonable limits. The dependence of the optimization function on exciton energy for different D/h ratios is presented in Fig. 1. For $D/h = 4$ and $D/h = 5$ the optimization function is nonmonotonous with a maximum at h =2.5 nm and h =2.0 nm, respectively. For larger D/h the largest value of optimization function is reached for the smallest dots among those investigated, with the height of h =1.5 nm. The most optimal dots emit in the range 3.2 – 3.8 eV, as can be seen in Fig. 1. Experimental results on single photon sources operating at 200 K reported in Ref. [7] show the emission energy of around 3.5 eV, and are in very good agreement with our theoretical predictions presented here. This also suggests that their QD geometry is most likely very close to an optimal one. It was reported in Ref. [7] that the estimated dimensions of the dots based on atomic force microscopy (AFM) measurements are: the height of 4 nm and the diameter of 25 nm. Our calculation for these dimensions of QDs yields an emission energy of 1.5 eV only, as well as very low values of the optimization function. However, AFM is a surface technique that measures the uncapped dots. Significant changes in the geometry of the dots after capping are possible and we believe that the dots measured in Ref. [7] actually have a much smaller height (i.e., reduced effective confinement region) than reported based on AFM measurements. We conclude with the discussion on the effect of the second order piezoelectricity [14] on the bound bi-excitons in the GaN/AlN wurtzite QD single photon sources.

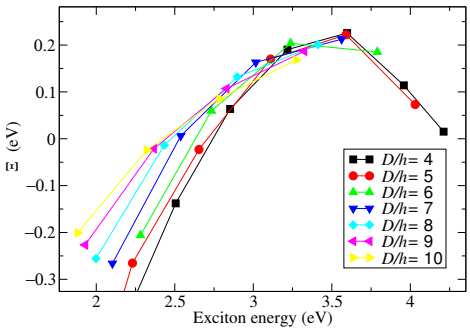

Fig. 1: The dependence of the optimization function defined as $\Xi(E_{\mathrm{X}})$ (see text for details) on exciton energy for different values of diameter to height ratios D/h.

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