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Size dependent biexciton binding energies in GaN quantum dots

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Single GaN/Al(Ga)N quantum dots (QDs) have been investigated by means of microphotoluminescence. Emission spectra related to excitons and biexcitons have been identified by excitation power dependence and polarization resolved spectroscopy. All investigated dots exhibit a strong degree of linear polarization ($\sim 90\%$). The biexciton binding energy scales with the dot size. However, both positive and negative binding energies are found for the studied QDs. These results imply that careful size control of III-Nitride QDs would enable the emission of correlated photons with identical frequencies from the cascade recombination of the biexciton, with potential applications in the area of quantum information processing. © 2011 American Institute of Physics. [doi:10.1063/1.3670040]

Individual photons created by the optical recombination of excitons confined in semiconductors quantum dots (QDs) have the potential for applications in the area of quantum information technology, including quantum cryptography^{1,2} and optical quantum computing.³ The asymmetry induced excitonic fine structure splitting (FSS) and the biexciton binding energy (E_{xx}^b) are the fundamental QD parameters of relevance for the possible generation of quantum entangled photon pairs in a cascade recombination of the biexciton.⁴⁻⁸ Both FSS and E_{xx}^b are parameters determined by the Coulomb interactions, which are related to the QD size and shape, as well as to internal or external fields such as electric, magnetic, and strain fields. As the FSS excludes entanglement of a photon pair, numerous reports have addressed various methods to tune and/or minimize the FSS of the QDs by applying external fields.^{7,9-13}

Recently, a photon time reordering scheme was proposed, providing an alternative path for obtaining entangled photons without the requirement of zero FSS.⁶ This scheme instead requires a vanishing E_{xx}^b , implying that the vertically (horizontally) polarized component of the biexciton emission has an identical photon energy as the horizontally (vertically) polarized component of the exciton. The control of E_{xx}^b by an external lateral electric field was proposed in a theoretical approach¹⁴ and demonstrated experimentally for InAsP QDs.⁵ However, for an electrically pumped photon source, the need of an additional external electric field for independent tuning of E_{xx}^b is an obvious drawback, requiring a complicated device design. A more straight forward approach, applicable to a wider class of QDs, is the control of the biexciton binding energy by an externally applied biaxial stress⁴ or by a direct control of the dot size.¹⁵

In this work, we report on the E_{xx}^b of GaN QDs, which is found to vary in a wide range, >12 meV, but it is also demonstrated to be either positive or negative essentially depending on the lateral size of the QDs. We have investi-

gated a GaN QD sample grown by molecular beam epitaxy on a c-plane sapphire substrate at a temperature of 720°C in the Stranski-Krastanov (SK) growth mode. A 5 nm AlN nucleation layer was grown on the substrate followed by a 90 nm GaN buffer layer and a 90 nm thick AlN barrier layer. The GaN QDs were formed from 6 monolayers of GaN, known to give rise to a bimodal size distribution of the QDs with a very low density of small dots (height ~ 1.6 nm and diameter ~ 10 nm) and a high density of large dots (height ~ 5 nm).¹⁶ The QDs were finally capped with a 50 nm $\text{Al}_{0.5}\text{Ga}_{0.5}\text{N}$ layer.

Microphotoluminescence (μPL) of single dots was measured with excitation at 266 nm from a continuous-wave laser. The linear polarization component of the PL emission was measured by a rotating a half-wave retardation plate in front of a fixed linear polarizer in the signal path.¹⁷

Only the small QDs are expected to provide sufficient electron-hole overlap to be optically active, with an estimated density of about $5 \times 10^8 \text{ cm}^{-2}$ as monitored in the recorded μPL spectra. Fig. 1(a) shows μPL spectra of a GaN QD (labeled Dot A) measured for different excitation powers (P_{ex}). Below the saturation limit of ~ 0.1 mW, the integrated intensities of peaks X and XX exhibit linear and quadratic power dependencies (see Fig. 1(b)), i.e., as expected for an exciton and a biexciton, respectively. In order to confirm that the peaks X and XX indeed originate from the same QD, the polarization dependencies of the peaks were analyzed (see Fig. 1(c)). For the same dot, the polarization directions are expected to be identical for X and XX, while the degrees of linear polarization are expected to be very similar.^{17,18} Since the integrated intensities of both X and XX in Fig. 1(c) were measured with an excitation power near the saturation limit of X, the intensity of X was fairly unaffected by variations in the optical pumping caused by small instabilities of the sample position within the excitation spot, while the intensity of XX was significantly more sensitive due to its quadratic power dependence. The peaks X and XX exhibit a strong polarization in the same direction with a similar degree of linear polarization, $P \sim 90\%$.¹⁹ Therefore, it is concluded

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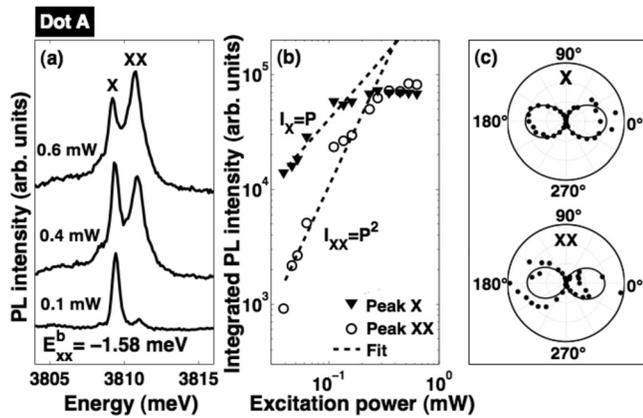


FIG. 1. μ PL data for Dot A. (a) Spectra as a function of the excitation power. (b) Integrated peak intensity as a function of the excitation power with fitted dashed lines. (c) Linear polarization dependence of the exciton and the biexciton with fitted solid lines.

that the peaks denoted X and XX likely are emissions related to the exciton and the biexciton from the same dot.

Fig. 2 shows the corresponding data for another QD (labeled Dot B), which reveals a positive biexciton binding energy, E_{xx}^b ($E_x - E_{xx}$) = 5.1 meV to be compared with -1.6 meV for Dot A. Moreover, the emission lines from different dots exhibit random polarization directions. The linear polarization is a result of the valence band mixing induced by the lateral anisotropy of the QD confinement potential.²⁰ This suggests random in-plane anisotropy of the QDs, which is consistent with the spontaneous QDs formation in the SK-growth mode. Any asymmetry of the confinement potential always result in finite FSS. However, no FSS could be resolved for any of the investigated QDs, probably due to a combination of the strong degree of linear polarization and relatively broad emission lines.¹⁷

The relative intensity of XX, with respect to X, increases with temperature (see Fig. 3(a)), which is attributed to an increased effective excitation of the QD at elevated temperatures. There is an increasing probability that charge carriers in shallow localization centers in the dot vicinity are released as the temperature is increased, to subsequently become trapped in the QD.

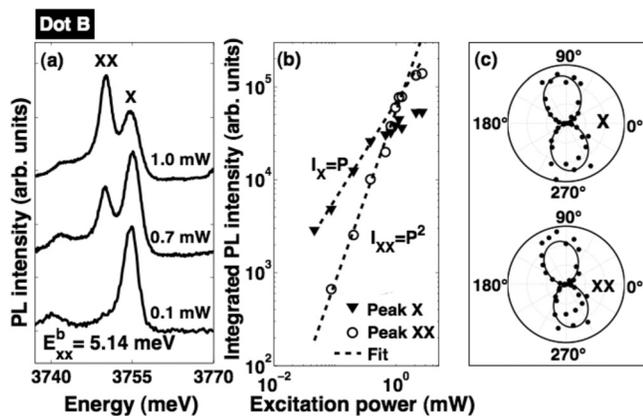


FIG. 2. μ PL data for Dot B. (a) Spectra as a function of the excitation power. (b) Integrated peak intensity as a function of the excitation power with fitted dashed lines. (c) Linear polarization dependence of the exciton and the biexciton with fitted solid lines.

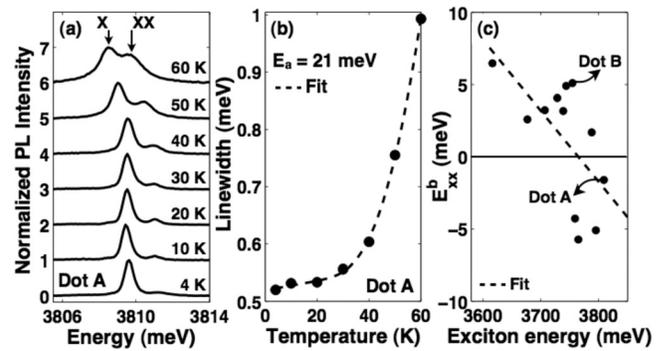


FIG. 3. (a) μ PL spectra as a function of temperature in the range 4–60 K for Dot A. (b) The exciton line width (dots) as a function of temperature with a fit according to Eq. (1) (dashed line) for Dot A. (c) The biexciton binding energy of 12 QDs as a function of the exciton emission energy. The dashed line is a linear fit to the experimental data and serves as a guide for the eye.

Fig. 3(b) displays the thermal broadening of the spectral line, with a full width at half maximum (FWHM) $\Gamma(T)$, increasing from 0.5 to 1.0 meV as the temperature has increased up to 60 K (for Dot A). The temperature dependence of the excitonic line width is expected to exhibit a linear dependence related to scattering with acoustic phonons in addition to an exponential component with activation energy E_A due to optical-phonon scattering,²¹

$$\Gamma(T) = \Gamma_0 + \gamma_p T + \gamma_a \exp\left(-\frac{E_A}{k_B T}\right), \quad (1)$$

with the coupling coefficients γ_a and γ_p , as well as the line width Γ_0 at 0 K. In GaN, the optical-phonon energies are large resulting in a negligible contribution to the broadening for $T < 80$ K from optical phonons. Instead the maintained exponential behavior of the broadening as observed in Fig. 3(b) has been explained in terms of dephasing due to thermal excitation of carriers from the QD into the surrounding barrier.²¹ The data presented in Fig. 3(b) can be fitted well by an acoustic phonon coupling constant $\gamma_p = 0.8 \mu\text{eV K}^{-1}$ and an activation energy of $E_A = 21$ meV. Thus, 21 meV can be taken as an estimate of the exciton localization depth in the studied QD. The obtained values of γ_p and E_A are comparable to what has been reported earlier for shallow InGaN QDs,²¹ but γ_p is here about one order of magnitude smaller than what was recently reported for significantly deeper GaN/AlN QDs.²² The shallow localization depth for the investigated GaN QDs is consistent with the asymmetric barrier with AlN below the QDs and $\text{Al}_{0.5}\text{Ga}_{0.5}\text{N}$ above. The barriers constitute a shallow confinement potential in the conduction band in the presence of a built-in electric field. Shallow potentials for the investigated QDs are further supported by the spectral absence of any emission related to excited states of the QDs.

The sample exhibits QDs with both positive and negative values on E_{xx}^b , as further exploited for 12 individual QDs summarized in Fig. 3(c). The biexciton binding energies were found to vary from 6.5 to -5.7 meV, exhibiting a trend of decreasing biexciton binding energy with increasing exciton emission energy. Note that this trend is opposite to what has been predicted theoretically for QDs modeled with a fixed shape, where E_x and E_{xx}^b depend on the dot size or

composition.^{23,24} A simple model for the biexciton binding energy is given by $E_{xx}^b = 2J_{eh} - J_{ee} - J_{hh}$,²³ where J_{eh} is the attractive electron (e) – hole (h) Coulomb energy, and J_{ee}/J_{hh} are the corresponding repulsive e-e/h-h energies, respectively. The strong built-in electric field across the GaN QDs separates the electrons and holes vertically, resulting in a reduction of J_{eh} and an enhancement of the repulsive interactions J_{ee} and J_{hh} . In particular, for thick QDs with well separated electrons and holes, the repulsive interaction energies dominate, leading to strongly negative biexciton binding energies.²⁵ Among QDs with equal heights, the vertical separation between e and h is approximately fixed, and the binding energy is then mainly dependent on the lateral confinement.²⁵ In QDs with successively smaller lateral dimensions, corresponding to increased exciton emission energies, the e-e and h-h interactions have also increased and resulted in reduced biexciton binding energies. Thus, the observed trend in Fig. 3(c) is explained in terms of different lateral confinement in QDs with approximately equal thickness. A similar trend for positive values on E_{xx}^b has previously been reported for GaN/AlN QDs.²⁵ The existence of both positive and negative binding energies in Fig. 3(c) demonstrates that it is possible to achieve vanishing biexciton binding energies for GaN QDs, also without external electric or stress fields.

It is notable that positive biexciton binding energies in GaN QDs are not well understood by means of the current approaches and material parameters used to determine the built-in electric field. The binding of the biexciton is essentially an effect of the Coulomb correlations, but the strong built-in electric field in GaN QDs results in large h-h and e-e repulsive interactions, which cannot be overcome by the attractive e-h interactions and the Coulomb correlations.²⁴ However, the Coulomb correlations are most significant in weakly confined QDs, and they can usually be neglected in the strong confinement regime.²⁶ The large experimental values of the biexciton binding energies up to 6.5 meV, as estimated for the GaN/Al(GaN) QDs in this work, can thus be associated with the shallow exciton confinement potentials of these QDs.

In summary, the exciton and biexciton in GaN QDs were spectrally identified by excitation power-dependence and optical polarization measurements. The degree of linear polarization was strong for all investigated QDs, but its angular orientation differs from dot to dot, suggesting random in-plane anisotropy of the QDs. The lateral dot size has a significant impact on the biexciton binding energy, resulting in an observed spread of the binding energies from –6 meV to +6 meV for the ensemble of measured QDs. These results demonstrate that the vanishing biexciton binding energies can be obtained for GaN QDs, without any external electric or stress fields. Thus, a careful control of the dot size would enable the fabrication of electrically pumped quantum light sources, emitting correlated photon pairs with

intrinsically identical photon energies for quantum information applications.

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