Excitonic complexes in InGaAs/GaAs quantum dash structures

A Musiał¹, G Sęk¹, P Podemski¹, M Syperek¹, J Misiewicz¹,
A Löffler², S Höfling² and A Forchel²

¹Institute of Physics, Wrocław University of Technology,
Wybrzeże Wyspiańskiego 27, 50-370 Wrocław, Poland

²Technische Physik, Universität Würzburg, Wilhelm-Conrad-Röntgen-Research
Center for Complex Material Systems, Am Hubland, D-97074 Würzburg, Germany

E-mail: anna.musial@pwr.wroc.pl

Abstract. Optical properties of single self-assembled InGaAs/GaAs quantum dashes have been investigated. The excitation power dependent measurements together with the rate equation model calculations enable to identify the excitonic and biexcitonic emission from one quantum dash. Based on that the biexciton binding energy and the relative exciton to biexciton lifetime ratio have been estimated. The emission spectra have also revealed a fingerprint of Coulomb interaction between excitons confined in the quantum dash and adjacent wetting layer. The conditions required for observing this phenomena and the influence of the wetting layer on the emission of individual quantum dashes have been discussed.

1. Introduction
Semiconductor quantum dash technology development is mostly driven by their optoelectronic applications. Quantum dash ensemble used as an active region in lasers or optical amplifiers exceeds in some cases the standard quantum-dot emission properties. This has already been proven for InAs dashes on InP and the improved device performance has been demonstrated [1].

Recently, the possible applications of quantum dashes have evolved toward the single-dash ones and quantum electrodynamics experiments (e.g. single photon sources or quantum bits for quantum cryptography and computation) as both the weak and strong coupling between excitons confined in single dashes and the three-dimensionally confined electromagnetic field was proven in InGaAs/GaAs material system [2]. This is possible due to relatively big sizes (from tens up to 100 nm in the elongation direction) and weakening of the quantum confinement resulting in enhanced oscillator strength of optical transitions. Those prospective results demand further and more comprehensive investigations of optical properties of individual quantum dashes, especially that, there is still a lack of both experimental data and theoretical calculations regarding such InGaAs dashes on GaAs. The existing reports have rather preliminary and qualitative character especially in excitonic complexes theme [3]. In this paper more systematic and quantitative investigations of InGaAs/GaAs quantum-dash structures is presented.
2. Experimental details

2.1. Investigated structures
All presented results concern the epitaxially (MBE) grown self-assembled structure with quantum dashes formed of nominally 4.5 nm In$_{0.3}$Ga$_{0.7}$As layer deposited on GaAs. Further growth details [4] and ensemble properties [5] can be found elsewhere. The indium content of about 30% enables exceeding the critical thickness for 3D island formation. Low-strain growth conditions results in formation of (5 x 30 x 100) nm structures preferentially elongated in one of the lateral directions ([1-10]). The surface density of quantum dashes is rather low - (6 – 9) $10^9$ cm$^{-2}$ resulting in very few quantum dashes to be excited within the laser spot. To further increase the spatial resolution a submicron mesa structures (from 1 µm down to 100 nm) were etched on the sample surface, providing 10 quantum dashes within smaller mesas, or less. The emission of individual objects have been additionally spectrally resolved due to size nonuniformity of self-assembled structures.

2.2. Experimental setup
Optical measurements have been realized in microphotoluminescence setup composed of microscope objective with high numerical aperture (0.4) and long working distance (20 mm), 0.5 m focal single grating spectrometer and InGaAs nitrogen-cooled CCD linear detector. Investigated structures were nonresonantly excited by a semiconductor laser emitting at 660 nm. Described setup characteristics (1 µm spatial and below 100 µeV spectral resolution) are sufficient for single-dot spectroscopy. The samples were mounted in a continues-flow microscopy-type cryostat and cooled with liquid helium down to the temperature of approximately 5K. For time-resolved photoluminescence setup details see ref. 6.

3. Results and discussion

3.1. Excitonic complexes
For single-dot investigations mesa structures sizes of hundreds nanometers have been chosen. They contain number of quantum dashes small enough to resolve single emission lines and at once the mesa sizes are sufficiently big to decrease the influence of their edges (e.g. stronger local electric field fluctuations due to the charges at the mesa sidewalls) broadening the emission. Exemplary microphotoluminescence spectra have been presented in figure 1. The individual emission lines both with the gaussian-type background (characteristic for the nonuniform quantum objects ensemble) due to relatively big mesa size (700 nm) can be clearly seen.

![Figure 1.](image_url) Low temperature normalized microphotoluminescence spectra of InGaAs/GaAs 30% In content quantum dashes for different excitation power values.

Observed lines are relatively broad (hundreds of µeV) compared to the setup resolution due to spectral diffusion effects – fluctuations of the charge environment in the time scale much smaller than the time of the measurements. Those effects are stronger for quantum dashes because of their relatively big sizes with respect to the mesa size causing the location of a structure in the vicinity of mesa edge more probable and hence stronger influence of the mesa sidewalls surface charge...
fluctuations. Choosing emission lines from the low energy tail of the spectra provides better separated lines originating from ground state emission. Higher energy part of the spectra revealed a wetting layer emission centered at 1.345 eV. Excitation power dependent measurements allow identification of excitonic and biexcitonic emission which intensity dependence is in low excitation conditions linear and quadratic, respectively. As the excitation power increases (above a critical value sufficient for biexciton formation) the excitonic emission saturates and the biexciton starts to dominate the spectra. To describe theoretically the emission intensity as a function of excitation power in the whole range of measured parameters three level rate equation model has been used [7,8]. The agreement between the modeled and measured dependences supports the origin of observed emission from one quantum dash. This allowed biexciton binding energy determination (as a spectral distance between excitonic and biexcitonic emission lines). For investigated structures it is in the range of 1 – 3 meV, which is consistent with our expectations regarding the strength of quantum confinement – due to their relatively big sizes they are rather in weak confinement regime. The best fit has been obtained for the exciton to biexciton lifetime ratio (being the only free parameter of this procedure) lower than 1 – see figure 2. This value is an additional fingerprint of the weak quantum confinement [9].

3.2. Sideband feature
Another very interesting feature in the emission spectra of individual quantum dashes under investigation is an undoubted asymmetry of biexcitonic line denoted as ‘sideband’ in figure 3. This low-energy emission band accompanying the biexcitonic line has been so far observed for monolayer fluctuation GaAs/AlGaAs quantum dots [10]. The sideband can be interpreted as a fingerprint of the Coulomb interaction between the biexciton and the excitons confined in the neighboring environment (e.g. wetting layer) which lowers biexciton emission energy. The energy difference between the ‘bare’ biexciton and interacting one is increasing with increasing number of interacting excitons. The sideband is composed of biexcitonic emission in different environmental conditions (different wetting layer occupation). As a result the microphotoluminescence measurements are probing not only the state of the quantum object but also the state of its surrounding, because the interaction with the environment cannot be separated or neglected. This phenomena can be observed only when the quantum dash (bi)excitons have lifetimes comparable with the wetting layer excitons. This condition guarantees that the Coulomb interaction is efficient. There are two possible cases: i) the emission lifetime of quantum dashes is very short or ii) the wetting layer states have localized character (like in [11]) and their lifetimes are longer than for typical quantum well. Time-resolved measurements confirmed that for investigated structures both is true, revealing relatively long radiative lifetime of excitons confined in the wetting layer (caused probably by localization effects) and short biexciton lifetimes (in agreement with predicted enhanced oscillator strength). Furthermore

![Figure 2.](image-url) **Figure 2.** Excitonic and biexcitonic emission lines intensity vs. excitation power analysis in double logarithmic scale: experimental points and theoretical curves obtained from rate equation model for fractional exciton to biexciton lifetime ratio.

![Figure 3.](image-url) **Figure 3.** Experimental emission spectra of single quantum dash for high enough excitation power to provide equal emission intensity of exciton and biexciton and appearance of low-energy sideband accompanying the biexcitonic line.
obtained results have shown that observed cases of biexcitonic emission accompanied by the sideband are limited to the higher energy part of the emission band (figure 4).

![Normalized relative sideband to biexciton emission intensity for different biexciton cases (defined by their emission energy) against the background of the photoluminescence spectrum of the quantum-dash structure.](image)

Moreover they differ in the intensity of the sideband with respect to the intensity of biexcitonic emission line. To compare those values for different quantum dashes integrated relative intensity has been obtained in rather high excitation regime providing almost equal excitonic and biexcitonic emission intensity. This could be a result of difference in (bi)exciton lifetimes or the efficiency of carrier capture between quantum dashes within the ensemble which tunes the experimental conditions out of the optimum for efficient Coulomb interaction.

4. Conclusions

Optical properties of single InGaAs/GaAs quantum dashes have been examined. Biexciton binding energy of about 1 – 3 meV has been obtained. The comparison of measured and modeled by rate equations excitation power emission intensity dependence enabled to indirectly estimate the kinetic properties of carriers confined in quantum dashes. Determined fractional exciton to biexciton lifetime ratio is an evidence of weak confinement regime in these structures.

The effect of lowering the biexciton emission due to Coulomb interaction with excitons confined in their neighborhood has been observed. The condition of comparable lifetimes of those excitations fulfillment has been confirmed by time-resolved measurements. As it has been shown in photoluminescence measurements one cannot separate the optical properties of investigated structure from its environment. In this sense this structure has to be treated as an open system.

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