Multi-excitonic structures in GaAs quantum dots proved by photon correlation spectroscopy

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Abstract. We studied micro-photoluminescence (µ-PL) spectra of a single GaAs self-assembled quantum dot (QD) using a pulsed excitation light source. At low excitation, we observed single photon emission from a neutral exciton in an isolated QD. For higher excitation, multiple peaks appeared in the spectra, reflecting the formation of exciton complexes. Cross-correlation functions between these lines showed either bunching or antibunching behavior, depending on whether the relevant emission was from a biexcitonic cascade or a charged exciton recombination.

1. Introduction
The quantum nature of light emitted from single semiconductor quantum dots (QD) has attracted much attention for realizing secure communications or ultra-fast parallel computations [1]. Production of single photons was demonstrated in a variety of QDs [2, 3]. A bosonic feature for indistinguishable photons emitted from the single photon source was identified [4]. Furthermore, the generation of entangled photon pairs associated with a biexciton-exciton cascade was proposed [5] and experimentally verified [6, 7]. So far, many studies for the generation of single and correlated photons have focused on (In,Ga)As QDs in an effort to develop a nonclassical photon source operating at near-infrared telecommunication wavelengths [8–10]. For practical application, however, it would also be desirable to work in shorter wavelengths where commercial silicon-based single photon detectors reach their maximum quantum efficiency. In this paper, we report on the generation of triggered single photons and correlated photons on self-assembly grown GaAs/(Al,Ga)As QDs emitting at a wavelength of 670 nm. Auto- and cross-correlation functions are analyzed for the emission of exciton complexes inside a single QD.

2. Sample Structure and Experimental Setup
The experiments were performed on GaAs self-assembled QDs in an Al₅ₓGa₇₋ₓAs barrier, grown by modified droplet epitaxy (MDE) [11]. Atomic force microscopy and high resolution scanning electron microscopy demonstrated the formation of QDs of 20 nm in height and 40 nm in base size, with a surface density of 7x10⁸ cm⁻². However, the mean size of QDs was reduced due to the interdiffusion of group III species during the post-growth annealing process of MDE-QD samples [12]. For
excitation, frequency-doubled output of a mode-locked Ti-sapphire laser was used. The laser produced 150 femtosecond pulses of 400 nm in wavelength and 76 MHz in repetition rate. A confocal microPL setup with an objective lens of 0.42 numerical aperture was used to capture individual QDs. The PL signal was split by a 50/50 chromium beamsplitter, both signals being coupled to single-mode optical fibers of 3.2 µm mean field diameter. Then, each beam was fed into a grating monochromator equipped with a silicon avalanche photodiode (APD). Electric pulses from the two APDs were sent to a time-correlated coincidence counter, each pulse acting as a start or stop event for the coincidence measurement of a Hamby, Brown and Twiss setup [13]. PL spectra were characterized using a N2 cooled charge-coupled-device. All experiments were performed at 8 K.

3. Results and Analysis
Figure 1(a) shows a typical time-integrated PL spectrum of the QD under a strong excitation condition (14 µW). At low excitation below 2 µW (not shown here), a single line, referred to as X, appears at 665 nm (1.8654 eV). This line is assigned by recombination of neutral excitons. Its linewidth is 0.45 meV in full width at half maximum (FWHM), being limited by instrumental resolution. As compared to bulk GaAs, the exciton transition energy is blue-shifted by ~350 meV due to the quantum confinement effect.

![Figure 1. A typical micro-hotoluminescence spectra of a single GaAs quantum dot under a strong excitation condition (14 µW).](image)

With increasing excitation intensity, several spectral components manifested themselves on the lower energy side of the line X. Here we will pay particular attention to the two bright lines referred to as CX and XX. The energy split between X and CX is 1.7 meV, and that between X and XX is 4.0 meV.

Excitation power dependence of the emission peaks are plotted in Fig. 2. All peaks increase with excitation power until they reach their saturation levels at ~15 µW. The dependence of peak X could fit to a power law with the exponent of 1.1 (±0.1), much larger than unity. The exponents of a power-law for lines CX and XX are evaluated to be 1.6 and 2.1 (±0.1), respectively. Since the value for peak XX is nearly double that of X, the formation of a two-exciton complex (biexciton) should appear in the emission of XX.

![Figure 2. Time integrated intensity as a function of excitation power for the three lines identified by X,CX,and XX in Fig. 1.](image)
photons were emitted synchronously with pulsed excitation of 76.0 MHz. The lack of a peak at zero time delay indicates that there is almost no probability of finding two or more photons inside each emitted pulse.

The results of cross-correlation measurement between peaks X and CX, X and XX are presented in Figs. 3(d) and 3(e), and those between peaks X and XX are shown in Fig. 3(f). The X–CX and X-XX correlations show the lack of a zero-time peak, similar to the X–X autocorrelation. In contrast, the X–XX correlation shows the emergence of a large zero-time peak.

Figure 3. Photon correlation histograms between X, CX, and XX emission lines for six combinations: (a) X-X, (b) CX-CX, (c) XX-XX, (d) X-CX, (e) X-XX, and (f) CX-XX.

The observation of an antibunching feature in the X–CX (XX-CX) correlation demonstrate that the CX and X (CX and XX) photons were not emitted simultaneously. This may be attributable to the line CX having originated from a recombination of charged excitons and the line X (XX) from neutral excitons (biexcitons). Since the transition between a charged exciton and a neutral exciton (biexciton) requires the injection or extraction of a carrier, the relevant process is much slower than recombination. As a result, either the X (XX) photon or the CX photon is generated during a single emission cycle, leading to the observation of the antibunching dip.

The bunching feature in the X–XX correlation suggests that line XX originated from biexcitons. When a biexciton is inside a QD, the first photon is emitted at the biexciton energy. Then, the second photon is emitted at the single exciton energy. This leads to the appearance of bunching behavior. The biexciton binding energy (~4 meV) is much larger than the bulk value (<1 meV), and almost reaches that of the exciton in bulk GaAs (4.9 meV).

To confirm the above explanation we took time-resolved measurements of the three emission peaks (Fig. 4). The data was fitted with mono-exponential decay applied to decay curve (as indicated by the solid lines in Fig. 4). The obtained decay times are 1.3±0.1, 1.1± 0.1, 1.0± 0.1ns for the X, CX, and XX states, respectively. We found that the peak XX appears and decays in the initial stage of the emission, then the peak X appears and eventually decays. This is a signature for the biexciton cascade results of cross-correlation measurements between X and XX [14].
Figure 4. Intensity of the X, CX, and XX photoluminescence as a function of time following the excitation pulse. The solid lines represent exponential decays.

4. Conclusion
We observed second-order correlation functions between emissions from exciton complexes in a single GaAs QD. Generation of single photons at a wavelength of 665 nm was confirmed. Both bunching and antibunching features were identified in cross correlation functions, revealing the origin of specific PL lines in a deterministic manner.

Acknowledgements
This work was supported by a 21st Century COE Program at Tokyo Tech “Nanometer-Scale Quantum Physics” by the Ministry of Education, Culture, Sports, Science and Technology, by Grant-in Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology of Japan.

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