Sequential Photon Emissions from Quad-excitons in Single GaAs Quantum Dots

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Abstract. We studied photoluminescence (PL) spectra of a single self-assembled GaAs quantum dot (QD) grown by modified droplet epitaxy. The single QD exhibits several sharp peaks in the PL spectrum indicating the formation of multi-excitonic complexes. Most of these peaks are originated from the recombinations of electron-hole pairs in the lowest level of the QD. We observed several emission peaks arising from recombinations from the second level of the QD at the condition of strong excitation. The difference of the recombination energy between the lowest and second levels is about 30 meV. We identified the emissions of tri-excitons and quad-excitons in the PL spectrum by single photon correlation spectroscopy. Quad-exciton, which is the four-exciton complex, emits correlated four photons in the cascade recombination. We observed bunching of photons emitted from the cascade by cross-correlation measurements.

1. Introduction
Semiconductor quantum dots (QDs) exhibit various interesting properties, which can not appear in bulk materials [1, 2]. Discretization of the energy structure and formation of multi-excitonic complexes, due to strong confinement effect, are one of the most attractive features for application into nano-scale electronic devices [3-5]. Multi-excitonic complexes are expected for a candidate of non-classical light sources [5]. The quantum features of light are essential to realize quantum computation and quantum cryptography [6]. In the single QD spectroscopy, the production of single photons has been performed by the recombination of excitons [7]. Biexciton cascade are demonstrated to generate correlated photon pairs [8]. Especially, it is shown that the photon pairs of biexcitons have entanglement as for their polarization in symmetric QDs [9, 10]. In practice, larger number of correlated photons are required for the quantum information processing [6]. Multi-excitonic complexes are suitable sources of large number of photons because they can emit photons depending on the number of excitons included in themselves. However, the spectra of QDs are complicated with increasing carriers because many-body effects, especially the exchange interaction, become pronounced [11, 12]. Therefore, understanding of the properties of few-particle states are essential to improve insight into the energy structure and the electronic properties of QDs.

In this report, we observed photoluminescence (PL) spectrum of multi-excitonic complexes recombining an electron and a hole in the second levels of a QD. The emission peaks of tri-excitons, which is the bound state of three excitons, and quad-excitons, which is the bound state of four excitons, are identified in the PL spectrum by single photon correlation measurements. The energy structure of the quad-exciton is discussed by considering the exchange interaction.
2. Experimental

Micro-PL measurements were performed on GaAs QDs grown by modified droplet epitaxy (MDE) [13, 14]. The GaAs QDs were formed self-assembly on a Al$_{0.3}$Ga$_{0.7}$As layer deposited on a GaAs substrate with [001] growth direction and capped by a Al$_{0.3}$Ga$_{0.7}$As layer. Atomic force microscopy and high resolution scanning electron microscopy demonstrated the formation of QDs with low density ($\sim 7 \times 10^8$ cm$^{-2}$), 20 nm in height, and 30 nm in base size before the capping. The size of the QDs is reduced due to interdiffusion of Ga and Al during post-growth annealing [15].

The QDs were kept at 8 K in a cryostat and excited by a diode laser, which produced 60 ps pulses of 407 nm in center wavelength and with 100 MHz repetition rate. A confocal microscope setup with an objective lens (NA = 0.55) was used for excitation and collection. The luminescence was separated by a beam splitter in a Hambury-Brown & Twiss (HBT) interferometer. Each beam was detected by an avalanche photodiode (APD) through a spectrometer equipping a N$_2$-cooled charge coupled device (CCD) camera. Outputs of the two APDs acted as start or stop events for a time-correlated coincidence counter, constructing a histogram of the measured time separation $\tau$. This setup can measure unnormalized second-order correlation function $g^{(2)}(\tau)$, which is useful to investigate the mechanism of the radiative decay in single QDs. When two states are excited exclusively each other in the same QD by the same excitation pulses, the cross-correlation function should show suppressed coincidence counts at $\tau = 0$ (antibunching), whereas if two photons are generated in a cascade process, the coincidence counts increase at $\tau = 0$ (bunching).

3. Results and Discussion

![Figure 1. Photoluminescence spectrum of a single GaAs QD. Two regions of the spectrum are displayed.](image)

Figure 1 shows a typical PL spectrum of a single QD. The spectrum has two groups of peaks separated by $\sim 30$ meV. In extremely low excitation power, no peaks are observed at the higher energy band 1.872 - 1.879 eV, while the peak $X$ appears at the lower energy band 1.842 - 1.852 eV. $X$ can be identified to originate from excitons, which emit photons by the recombination of electron-hole pairs in the lowest energy levels of the QD ($1e$ and $1h$). As the excitation power is increased, the intensity of $X$ increases linearly and several peaks appear at the low energy side of $X$. Their intensities increase superlinearly, indicating the formation of multi-excitonic complexes in the QD. The energy differences of each peak depend on their binding energies. In previous works, we have studied the origin of PL peaks of the QDs by polarized PL, time resolved PL, excitation power dependence, and photon correlation measurements. It is shown that neutral and positively charged excitonic complexes can be excited in the QDs, and the recombinations of $1e-1h$ pairs contribute to the peaks in the lower energy band [16, 17]. On the other hand, increasing the excitation power furthermore, several peaks appear at the higher energy band. Their intensities increase superlinearly. By considering the excitation processes, the origin of the higher energy band can be interpreted as the emissions from the second levels in the QD ($2e$ and $2h$). In this experiment, carriers are excited in the barrier at 3.05 eV and captured into the QD at 1.88 - 1.85 eV with energy relaxation. When the excitation power is
Coincidence Counts
Delay Time (ns) Delay Time (ns) Delay Time (ns)
(a) 2X - X (b) 3X - 2X (c) 4X - 3X

Figure 2. Results of cross-correlation measurements for neutral excitonic complexes.

low, the probability to find one or two excitons in the QD is dominant so that the recombinations of 1e-1h pairs mainly occur. In such condition, carriers immediately relax into the lowest levels even if they occupy the second levels. In contrast, because the probability to find more than two excitons in the QD has a large value with increasing the excitation power, the lowest levels are fully occupied and the recombinations of 2e-2h pairs begin to take place. Thus, the higher energy band should be attributed to highly multi-excitonic complexes.

We assumed 3X and 4X in Fig. 1 as the emissions from 2e-2h pairs in tri-excitons and quad-excitons, respectively. The tri-exciton consists of two electrons and two holes in the lowest levels and one electron and one hole in the second levels \((1e^21h^22e^12h^1)\), where the superscripts denote the numbers of carriers occupied in the levels. The quad-exciton has four electrons and four holes, and the lowest and the second levels are fully occupied \((1e^21h^22e^22h^2)\). If the peak 4X results from the quad-excitons, correlated four photons should be emitted by the cascade recombination which involves tri-excitons, bi-excitons, and excitons. To confirm this assignment, we performed single photon correlation measurements.

There are many peaks without labels in the PL spectrum. These peaks are originated from charged excitonic states because each of them shows antibunching in the cross-correlation with each neutral state. We do not mention furthermore about the charged states in this report. The intensity of the charged states are much larger than that of neutral states because the sample has slightly p-doped structure introduced not intentionally by the growth process \[18\].

The results of cross-correlation measurements are displayed in Fig. 2. As shown in Fig. 2(a), the cross-correlation between 2X and X shows apparent bunching in the central peak at \(\tau = 0\), which corresponds to the probability that both X and 2X are emitted during the same laser pulse. The visibility of the bunching in 2X-X is a higher value than other cross-correlation measurements associated with X. Therefore, 2X can be assigned as the biexciton emission.

The cross-correlation function between 3X and 2X is presented in Fig. 2(b). Since bunching is observed at \(\tau = 0\), 3X and 2X are emitted in the same cascade process. This indicates that 3X is the tri-exciton emission. When the 2e-2h emission of the tri-exciton occurs, the biexciton is left behind in the QD and then 2X and X can be emitted in order. The cross-correlation between 3X and X also shows bunching as supporting this assignment (not shown here).

Figure 2(c) is the result of cross-correlation between 4X and 3X, which shows bunching at \(\tau = 0\). In addition, 4X exhibits bunching in the cross-correlations function with other neutral states (not shown here). Therefore, 4X can be identified to the emission of quad-excitons. When the 2e-2h emission of the quad-exciton occurs, the tri-exciton is left behind in the QD and the cascade of the tri-exciton should take place. It is demonstrated from the above data that the temporally correlated four photons are emitted from the QD.

The energy structure of the second levels of the QD is important to understand the PL spectrum of the quad-excitons because the second levels have p-orbital-like envelop functions, where \(p_x, p_y, p_z\) are degenerate in a fully symmetric QD. If the degeneracy of the \(p_x, p_y, p_z\) is
retained or the energy spacings between them are smaller than the exchange energy between 2e-2e or 2h-2h, the ground state of the quad-exciton contains two electrons (holes) with spin-parallel in the lower two p\(_e\)-(h)-orbitals. (The accurate value of the 2e-2e or 2h-2h exchange energy could not be obtained, though these values would have a few meV because the exchange energy between 1e-2h is ~ 1.8 meV in the QD measured by an energy separation of emission peaks of the positively charged biexcitons [19].) On the other hand, if the energy spacing of lower two p\(_e\)-(h) orbitals is larger than the 2e-2e (2h-2h) exchange energy due to the anisotropic shape or strain of the QD, the stable condition for the quad-exciton is that two electrons (holes) occupy the lowest p\(_e\)-(h) orbital with anti-parallel spins. In the former case, 4X should have several peaks in the PL spectrum due to the exchange interaction [20]. However, we can not find any other peaks satisfying the properties of the quad-exciton: it have only one peak in the higher energy band. Therefore we adopt the latter case for the QD.

In conclusion, we observed the PL from the single GaAs QD arising from the emissions from the lowest levels and the second levels. It is shown that the quad-excitons are excited in the QD and several peaks emitted in the cascade recombination of the quad-excitons are identified in the PL spectrum by using the single photon correlation spectroscopy. The results demonstrate that the correlated four photons are emitted from the quad-excitons, where the two electron-hole pairs in the second levels recombine at first and then the recombinations of the two electron-hole pairs in the lowest levels take place.

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