# Spin-relaxation Dynamics of Excited Trion States in an InAs Quantum Dot

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## 1. Introduction

Semiconductor quantum dots (QDs) have been attracting many researchers because of its potential application to solid state quantum information devices. A single electron/hole spin of charged exciton (trion) state in a QD can be fairly suitable because of its long coherence time, which arises from deep confinement of a QD, as well as ease of preparation and detection of a QD trion by optical means. From this point of view, investigating spin dynamics in a single QD is essential for future device application.

In this paper, we demonstrated photon-correlation based analysis of spin dynamics of QD trions. We evaluated spin-relaxation rates of excited positive trions with the aid of a rate equation simulation. We also prepared another sample with rapid thermal annealing (RTA) [1], and found that their spin-relaxation rates were reduced compared with those without RTA.

## 2. Sample and experimental setups

### Sample

The QD wafer was grown by molecular beam epitaxy on a (001) GaAs substrate. The areal density of the QDs was estimated to be  $\sim 10^9$  cm<sup>-2</sup>. The trion recombination energy is tuned around 1.3 eV at 10 K by means of In-flush method [2]. Details of the growth condition can be found elsewhere [3]. We prepared two samples without and with RTA process for comparison, namely, QD A and B, respectively. For QD B, RTA was performed at 780 °C for 10 s. In order to excite only a single QD, we fabricated mesa structures of  $\phi \sim 1 \mu m$  for each sample.

# Experimental setups

Samples were cooled below 10 K in a liquid He flow cryostat. We used a continuous wave Ti:Sapphire laser (E  $\sim$  1.55 eV) for excitation and PL signals were analyzed by a monochromator with a Si multi-channel detector. Photon cross-correlation measurements were performed by using a Hanbury Brown-Twiss intensity interferometer.

#### 3. Results and discussions

## PL spectra

Figure 1 (a)[(b)] shows an observed PL spectrum for QD A [B]. Three prominent peaks are clearly seen for QD

A. We identified these peaks as optical radiation from the positive trion  $(X^+)$  and the positively charged biexcitions  $(XX^+s)$  based on time resolved PL measurements and excitation power dependence of PL intensities (not shown). For QD B, two other peaks as well as  $X^+$  and  $XX^+$  peaks are observed.



Fig. 1 (a)[b] Observed PL spectrum for QD A [B].

## Energy diagram

Figure 2 shows the corresponding energy diagram. One of the electron-hole (e-h) pair in a ground *s*-shell of  $XX^+$ state recombines, which leaves behind the other e-h pair in an *s*-shell and a hole in a *p*-shell (excited trion state:  $X^{+*}$ ). The  $X^{+*}$  consists of three half-integer unpaired spin, which leads to four doubly degenerated sublevels ( $X_1^{+*}...X_4^{+*}$ ) due to the e-h and hole-hole (h-h) exchange interactions [4,5]. For the successive decay of  $XX_i^+$  and  $X^+$ , the intermediate  $X_2^{+*}$  and  $X_3^{+*}$  states should be relaxed to ground  $X^+$  state via  $X_4^{+*}$  state. Their transition rates of  $\gamma_2$ ,  $\gamma_3$  and  $\gamma_4$  depend on the spin configurations. We supposed that  $\gamma_4$  is very high (>100 GHz) because the *p*-shell hole rapidly relaxes into the *s*-shell without changing its spin state [6]. Estimation of  $\gamma_2$  and  $\gamma_3$  is our purpose.

# Photon correlation measurements

In order to investigate the carrier relaxation dynamics, we performed photon cross-correlation measurements between  $XX_i^+$  and  $X^+$  PL peaks. Figure 3 (a) and (b) [(c) and (d)] show correlation function  $g^{(2)}(\tau)$  for  $XX_3^+ \cdot X^+$  and  $XX_2^+ \cdot X^+$  observed for QD A [B], respectively. For QD A,



Fig. 2 Schematic energy diagram for  $XX^+$ ,  $X^+$  and  $X^{+*}$  states.

we observed bunching behavior for  $XX_3^+ \cdot X^+$  at positive delay  $\tau > 0$ , which was not observed for  $XX_2^+ \cdot X^+$ . This indicates that  $\gamma_3$  is much higher than the radiative decay rate of  $X^+$  ( $\Gamma_X$ ) and that  $\gamma_2$  is comparable to or lower than  $\Gamma_X$ . By comparing the numerically simulated  $g^{(2)}(\tau)$  (solid curves in Fig. 3),  $\gamma_2$  is estimated to be 0.8-1.0 GHz, which is about one order of magnitude lower than  $\gamma_3$  (> 8 GHz). This behavior can be understood in terms of the spin configurations. For  $X_2^{+*}$  state, spin-flip of the *p*-shell hole is needed for transition to  $X_4^{+*}$  state. In contrast, only relative phase change of the *p*-shell hole is needed for the case of  $X_3^{+*}$  state. The former has less likelihood to occur than the latter because spin along the QD quantization axis is usually the most stable.

Meanwhile, the situation is different for QD B. We could observe only faint bunching behavior even for the  $XX_3^+$ - $X^+$  case. For QD B, we evaluated rates  $\gamma_{2(3)}$  of 0.3 (0.9) GHz, which are lower than those for QD A. The RTA process can reduce the number of defects inside and nearby the QD, which is one of the possible reasons of the reduced spin-relaxation rates.

The measured PL spectra also support our estimation of  $\gamma_2$  and  $\gamma_3$ . In Fig. 1(b), a set of two peaks (labeled  $X_2^{+*}$  and  $X_3^{+*}$ ) are clearly seen at ~1.38 eV, and their energy difference is exactly the same as that between  $XX_2^+$  and  $XX_3^+$ . The origin of the two peaks is the optical transition from  $X_{2(3)}^{+*}$  state to  $0^{+*}$  state without *p*- to *s*-shell hole relaxation [7]. This result leads to  $\gamma_{2(3)} \leq \Gamma_X$  for QD B. In contrast,  $X_2^{+*}$  and  $X_3^{+*}$  are very weak or below noise level for QD A [see Fig. 1(a)], which leads to  $\gamma_{2(3)} \geq \Gamma_X$ .

## 4. Conclusions

We demonstrated photon-correlation based analysis of spin dynamics of the excited trions in InAs single QDs. We



Fig. 3 Cross-correlation functions for  $XX_i^+-X^+$ .

evaluated the spin-relaxation rates for different spin configuration by comparing the experiments with the numerical simulations. We also found that the rates can be reduced with the RTA process. Our method and obtained results are very useful for realizing quantum information devices using spin states of a single QD.

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