Correlated photon emission in a thick barrier coupled quantum dot

Shohgo Yamauchi, Amane Shikanai, Isao Morohashi, Shigenori Furue, Kazuhiro Komori, and Takeyoshi Sugaya
National Institute of Advanced Industrial Science and Technology (AIST), 1-1-1 Umezono, Tsukuba, Ibaraki 305-8568, Japan and CREST, Japan Science and Technology Corporation (JST), 1-6-1 Takezono, Tsukuba 305-0002, Japan
Toshihide Takagahara
Department of Electronics and Information Science, Kyoto Institute of Technology, Matsugasaki, Sakyo, Kyoto 606-8585, Japan

(Received 10 June 2007; accepted 5 September 2007; published online 2 November 2007)

Correlated photon emission from a thick barrier coupled quantum dot (QD) has been observed by using selective two-color excitation spectroscopy and second-order photon correlation spectroscopy. Surprisingly, the carrier creation in both QDs induced an anomalous increase in the luminescence intensity, and furthermore the cross photon correlation spectrum between two QDs exhibited photon antibunching with a long recovery time. These significant findings can be interpreted in terms of the electromagnetic interaction between QDs with a thick barrier layer. © 2007 American Institute of Physics. [DOI: 10.1063/1.2802295]

I. INTRODUCTION

Compound semiconductor quantum dots (QDs) have long been studied, mainly in terms of their application as active layers in optical devices using their unique distribution of state density originating from three-dimensional quantum confinement. In recent years, single QD and coupled QD (CQD) systems have attracted attention as media for quantum information processing devices, because they maintain a single quantum state that can be controlled with ultrafast optical techniques and they have a high degree of affinity with optical communications systems. In fact, Li et al. have already demonstrated a quantum logic gate using the exciton-biexciton correlation in a single GaAs/AlGaAs QD. Furthermore, an entangled photon pair emitter has been proposed that consists of a CQD. A CQD system is considered essential for these quantum information devices in terms of realizing a scalable device configuration in which it is expected that the QDs constituting a CQD will have to be independently controlled. However, at present, the physical property of a CQD is less well understood. In CQD systems, one of two coupling mechanisms, namely, quantum mechanical coupling or electromagnetic coupling, will dominate depending on the interdot spacing. In general, quantum mechanical coupling exhibits exponential decay as a function of the interdot spacing, whereas electromagnetic coupling, typically dipole-dipole coupling, exhibits at most a power-law dependence on the interdot spacing and has a longer range than quantum mechanical coupling. In addition, unlike electromagnetic coupling, quantum mechanical coupling will prevent the control of individual QDs because of the wave function sharing between two QDs. Although there have been several experimental reports on CQD systems, the distinction between the quantum mechanical and electromagnetic coupling systems remains a matter for discussion.

In this paper, we discuss the correlated photon emission in an electromagnetic CQD by using selective two-color excitation spectroscopy and second-order photon correlation spectroscopy. A photon correlation experiment is well known to be a powerful technique for analyzing the competitive nature of exciton emissions in a QD. Although Gerardot et al. have already discussed the photon statistics in a CQD, there is a fear that the very thin barrier between the QDs used in their report induced strong quantum mechanical coupling and made it difficult to analyze the photon emission processes. By contrast, the CQD used in this report has a thick barrier layer, which effectively reduces quantum mechanical coupling and enables the individual excitation of two QDs. This allows us to discuss the electromagnetic interaction and the photon emission process in a CQD more clearly.

II. COUPLED QD SAMPLE

Our sample was a self-organized InAs QD embedded in a GaAs matrix grown by molecular beam epitaxy (MBE) with the indium-flush method. This sample was the same as that used in our previous work. Using the stacking growth technique, we obtained a CQD sample where two QDs were joined in the growth direction with an arbitrary barrier thickness. In this study, the barrier thickness d of the CQD is 7 nm, which weakens the quantum mechanical coupling sufficiently for us to disregard the carrier transfer between QDs. A single QCD was observed through an aluminum aperture mask (0.2 – 0.5 μm) fabricated on the sample surface by electron-beam lithography and lift-off technique. For the optical measurements, the sample was cooled to 6 K in a liquid helium cryostat, and excited by a continuous-wave tunable Ti:sapphire laser.

Figure 1(a) shows the microphotoluminescence (μPL) and photoluminescence excitation (μ-PLE) spectra from two pairs of CQDs. CQDs A and B were located in different aperture mask holes of the sample. The two PL peaks in the
spectra were assigned to two QDs (QD1, QD2) constituting a CQD. In our previous work, we described these two PL peaks as bonding and antibonding states. In this work, it may be better to express these peaks as the different QDs constituting a CQD, because the carrier transfer is negligible in d = 7 nm CQDs. We also mention here that the two peaks are less likely to be attributed to a charged exciton, judging from the discrepancy between the two PLE spectra and the polarization dependence of the PL peaks. A detailed discussion of PL peak assignment has been provided elsewhere. The PLE spectra in Fig. 1(a) show the discrete excited levels arising from three-dimensional quantum confinement in the zero absorption region (1.342–1.355 eV for CQD A). These excited levels vary between QD1 and QD2 as regards the reflection of the different luminescence energies, and this ensures that carrier transfer between QD1 and QD2 is prevented. Consequently, we can selectively excite one QD of a single CQD.

III. TWO-COLOR EXCITATION SPECTROSCOPY

A. Experiment and results

As shown in our previous report, the coexcitation of both QD1 and QD2 results in a noticeable phenomenon related to the interdot interaction. Figure 1(b) shows the PL spectra of the CQDs with one- or two-color excitation. With CQD A, while excitation at E1 (E2) yields only a QD1 (QD2) emission [bottom (middle) of the figure], two-color excitation (both E1 and E2) induces a noticeable increment in the PL intensity that can reach five times the intensity obtained in one-color excitation (top of the figure). This is a typical phenomenon for weakly CQDs (d = 7 nm), and exactly the same feature was observed for CQD B.

Figure 2(a) shows the excitation power dependence of integral PL intensities in CQD A. QD1 and QD2 were excited at E1 and E2, respectively. With two-color excitation, the detection of QD1 was performed under various E2 intensities with constant E1 excitation, and vice versa. The solid lines are curves fitting the exponent m to the excitation power. (b) Diagram of excitation condition and carrier occupation in a thick barrier CQD.

\[ \Delta I_{QD1} = I_{QD1}^{EC} - I_{QD1}^{EC}, \]
citation power. As can be seen, $\Delta I_{QD1}$ shows close to linear dependence on the QD2 excitation power ($m=0.8$).

**B. Theoretical model and interpretation**

The surprising increment in the PL intensity with two-color excitation presented in Fig. 1(b) cannot be explained by carrier transfer through the tunneling process (Dexter type energy transfer)\cite{28} or Förster type energy transfer.\cite{29} This is because one-color excitation would yield both QD1 and QD2 emissions if these processes occurred. Furthermore, the increment in the PL intensities is too large to be explained by the two-photon absorption process and the super-radiance effect.\cite{30} On the other hand, the linear dependence of $\Delta I_{QD1}$ on the QD2 excitation power presented in Fig. 2(a) reflects the fact that the PL increment is derived from the coexcited state $|QD1\rangle|QD2\rangle$ in which both QD1 and QD2 are excited, i.e., this is the transition of $|QD1\rangle|QD2\rangle \rightarrow |QD2\rangle$. In addition, the fact that the PL exhibits a large increment more than threefold indicates that the ratio of the PL intensity increase is very large.

Here, with our proposed model, this phenomenon originates from the enhancement of the intraband energy relaxation caused by the electromagnetic interaction between two QDs.

To consider this interpretation, we analyzed the increasing ratio of the PL intensity with two-color excitation. We consider the QD1 rate equation [see Fig. 3(a)]:

$$\frac{dN_2}{dt} = -\gamma_{21}N_2 + \Gamma N_0,$$

$$\frac{dN_1}{dt} = -\gamma_{10}N_1 + \gamma_{21}N_2. \quad (2)$$

From these equations, the populations of the lowest excited level $N_1$ and the second excited level $N_2$ are written as

$$N_1 = \frac{\Gamma \gamma_{21}}{\gamma_{10} + (\gamma_{10} + \Gamma) \gamma_{21}}, \quad (3)$$

$$N_2 = \frac{\Gamma \gamma_{10}}{\gamma_{10} + (\gamma_{10} + \Gamma) \gamma_{21}}, \quad (4)$$

where $N$ is the total number of carriers in this system. When the intraband energy relaxation $\gamma_{21}$ is assumed to be modulated by the carrier population of QD2, the increasing ratio of the PL intensity with two-color excitation is derived as

$$\frac{N_{2c}}{N_{1c}} = \frac{\gamma_{10}(1 + \frac{\gamma_{10} + \Gamma}{\gamma_{21}}) \gamma_{21}^{c_{2c}}}{\gamma_{10} + (\gamma_{10} + \Gamma) \gamma_{21}^{c_{21}}} \gamma_{21}^{c_{21}}, \quad (5)$$

where $N_{1c}$ and $N_{2c}$ are the carrier populations at the $N_1$ level for one- and two-color excitations, respectively, and $\gamma_{21}^{c_{1}}$ and $\gamma_{21}^{c_{2}}$ are the intraband energy relaxation rates for one- and two-color excitations, respectively. Here, we assume that the intraband energy relaxation $\gamma_{21}^{c_{21}}$ is proportional to the QD2 excitation power $P_{E2}$:

$$\gamma_{21}^{c_{21}} \propto N_{QD2} \propto P_{E2}.$$ \quad (6)

We analyzed the observation results based on these equations. Figure 3(b) shows the increasing ratio of the PL intensity $N_{2c}/N_{1c}$ as a function of the QD2 excitation power. The different symbols in the figure denote different QD1 excitation conditions. As shown in the figure, the fitting curves derived from Eq. (5) reproduce the observation results even under different QD1 excitation conditions ($\Gamma=0.42, 0.67$). Thus, this analysis strongly supports our interpretation that the PL increment with two-color excitation arises from the acceleration of the intraband energy relaxation caused by the interdot interaction. When we adopt $\gamma_{10}=1.24$ ns,\cite{27} we obtain $1/\gamma_{21}^{c_{21}}=5.2$ ns from the fitting parameters. Furthermore, we can estimate that the energy relaxation time $1/\gamma_{21}^{c_{21}}$ is reduced from 5.2 ns to 650 ps with the coexcitation of QD1/QD2 (at $P_{E2}=1$ mW).

**C. Discussion**

Here, we mention another possible way to interpret our observations: namely, that the defect levels near the QD are occupied by the photogenerated carriers in QD2, which prevents optical quenching, and consequently the QD2 emission is increased. We verified this interpretation with the following rate equation:

$$\frac{dN_2}{dt} = \Gamma N_0 - (\gamma_{21} + \gamma_{dd})N_2; \quad \frac{dN_1}{dt} = \gamma_{21}N_2 - (\gamma_{10} + \gamma_{dd})N_1; \quad (7)$$

where $\gamma_{dd}$ is the relaxation rate through the defect level. We assumed $\gamma_{21}^{c_{21}}=\gamma_{dd}/P_{E2}$, and evaluated $N_{2c}/N_{1c}$. However, we could not reproduce the experimental results with realistic
fitting parameters on the basis of Eq. (7): we needed $\gamma_d \gg \gamma_0$ (one order of magnitude) for a suitable fitting ($1/\gamma_d \sim 100–200$ ps). This kind of fast detection time in the defect level is unlikely because the nonradiative recombination time was estimated at $1–10$ ns in III-V compound semiconductor nanostructures.31,32

In the first place, if the defect level has an effect on our observations, the two-color excitation of QD1 will induce a phenomenon similar to the PL increment on the coexcitation of QD1/QD2. However, our observations using two-color excitation spectroscopy refute this prediction. In a previous report,20 we have shown the two-color excitation PLE spectrum of QD1. In that case, the QD1 was excited at two different excited levels by two lasers. In this case, if the above prediction of the defect level effect is correct, it might be expected that coexcitation at these excited levels would induce a large PL increment. However, the fact that there were no large PL increments conflicts with the above-mentioned assumption of a defect level effect.

From the above discussions, our interpretation of the acceleration of the intraband energy relaxation with the interdot interaction is thought to be the most likely reason for our observations. We suppose that the quasibicexcitonic state induced by the dipole-dipole interaction between excitons of QD1/QD2 allows extra rapid relaxation paths, which enhance the intraband relaxation per unit time, resulting in an increment in the PL intensity. The long intraband energy relaxation time evaluated at 5 ns may come from the triplet states in a QD. In passing, we observed no PL energy shift as a result of the two-color excitation in our experiments. This implies that the dipole-dipole interaction in $d=7$ nm CQDs is not strong enough to cause an energy shift larger than the spectral resolution (20 $\mu$eV).

IV. SECOND-ORDER PHOTON CORRELATION SPECTROSCOPY

A. Experimental procedure

Next, we undertook a further investigation of the correlated photon emission in a thick barrier CQD by using second-order photon correlation spectroscopy. First, we confirmed the single photon emission from each QD constituting the CQD. Figure 4(a) shows the experimental system we used for time correlated photon counting spectroscopy pursuant to the Hanbury-Brown-Twiss setup.33 Two monochromators select photons from an arbitrary QD (QD1 or QD2), which are then fed into single photon detectors (Si avalanche photodiode). By using start and stop signals from the detectors, the time correlated counting board outputs the second-order photon correlation spectrum:

$$g^{(2)}_{\alpha\beta}(\tau) = \langle I_{\alpha}(t)I_{\beta}(t+\tau)/\langle I_{\alpha}(t)\rangle \langle I_{\beta}(t+\tau)\rangle \rangle,$$

where the suffix $\alpha\beta$ indicates the selected QD as a start (stop) signal. Figures 4(b) and 4(c) show the autocorrelation spectra of QD1 and QD2, which constitute CQD B, with one-color excitation, where the wavelengths of both monochromators were set to that of QD1 or QD2. The two QDs were individually excited at $E_3$ or $E_6$ energy indicated in Fig. 1(a); this means that the interdot interaction between QD1 and QD2 was miniscule. Consequently, we can regard these observations as a result from a single QD. As shown in the figures, the autocorrelation spectra of both QD1 and QD2 ($g^{(2)}_{11}$ and $g^{(2)}_{22}$) exhibit dip structures around the delay time $\Delta t=0$. These structures originate from photon antibunching, which is evidence for the single photon emission from each QD constituting the CQD, as described in previous reports.31

B. Experimental results

1. Photon statistics with one-color excitation

In Fig. 5, we show the autocorrelation spectra of QD2 with one-color excitation whose energy was higher [$E_6$: Fig. 5(a)] or lower [$E_3$: Fig. 5(b)] than the excited level resonated with the longitudinal optical (LO) phonon $\Delta E_{LO} = 34.4$ meV, see Fig. 1(a)]. Whereas both spectra in Fig. 5 show antibunching at $\Delta t=0$ reflecting the single photon emission, the excitation at an energy lower than the LO-phonon level causes a bunching feature whose decay time $\tau$ is about 4 ns. This result indicates that the photon emission is stimulated during $\Delta t<\pm 4$ ns.

2. Photon statistics with two-color excitation

Figure 6 shows the photon correlation spectra of CQD B with two-color excitation, where QD1 (QD2) was excited at $E_3$ ($E_6$) energy indicated in Fig. 1(a). Figures 6(a) and 6(b)
show the autophoton correlation spectra of QD1 and QD2, respectively. Both spectra $g^{(2)}_{11}$ and $g^{(2)}_{22}$ exhibit the bunching and the antibunching features as found with the result in Fig. 5(b), where the bunching decay times differ from that in Fig. 5(b) because of the different excitation conditions (excitation power and two-color excitation). Figure 6(c) shows the cross photon correlation spectrum $g^{(2)}_{12}$ between QD1 (for a start signal) and QD2 (for a stop signal). It is noteworthy that spectrum $g^{(2)}_{12}$ exhibits a wide antibunching structure without bunching.

C. Theoretical model and interpretation

1. Photon emission process of one-exciton system in a CQD

In our thick barrier CQD, the photon statistics with selective one-color excitation will reflect the difference in the energy relaxation process in one QD of a single CQD. With an excitation energy higher than the LO phonon energy, the intraband energy relaxation of the photogenerated carriers will be rapid due to the strong exciton–LO-phonon interaction. On the other hand, when the excitation energy is lower than that of the LO phonon, the intraband relaxation of the carriers depends only on the acoustic phonon emission, which requires a time of several nanoseconds. The bunching feature in Fig. 5(b) indicates that the photon emission is stimulated during the bunching time when the intraband energy relaxation is caused by the acoustic phonon.

When the intraband relaxation is caused by the LO phonon ($E_0$ excitation), the cycle time of the exciton emission in a QD is considered to be roughly the exciton lifetime (~1 ns) due to the rapid energy relaxation process, and consequently the probability of detecting the next exciton emission is regarded as being constant in the DT > ±1 ns time domain. Therefore, $g^{(2)}_{12}$ will not exhibit bunching [see Fig. 5(a)].

On the other hand, when the carrier energy relaxation is caused by the acoustic phonon ($E_4$ excitation), the first exciton (detected as a start signal) will leave several acoustic phonons in a QD before the photon emission. In this case, it is just conceivable that these phonons remain in the QD for a while after the photon emission. Consequently, the second exciton generated in the QD (detected as a stop signal) will be scattered by these residual phonons (see Fig. 7). The phonon scattering rate is proportional to the phonon number, according to following formula:
where \( n_q \) is the number of phonons, \( a_q \) and \( a_q^\dagger \) are creation and destruction operators, and \( k \) and \( q \) are exciton and phonon wave numbers. The high scattering rate accelerates the carrier energy relaxation, and accordingly the probability of detecting the stop signal during the phonon survival time is expected to become higher than that in the DT \( \gg 0 \) time domain. On the basis of the above discussion, our observation of the bunching for DT \(< \pm 4 \) ns in Fig. 5(b) is considered to originate from the enhancement in the intraband energy relaxation caused by the residual phonons emitted from the start signal exciton. The decay time of the bunching (\(~\sim 4 \) ns) indicates the phonon dissipation time. Here, let us compare the bunching decay time with the phonon dissipation time estimated using classical statistical mechanics. It is well known that the phonon dissipation time is classically given by

\[
\tau = \frac{3\kappa}{C_v v^2},
\]

where \( C_v \) is the specific heat at a constant volume, \( \kappa \) is the thermal conductivity, and \( v \) is the phonon (sound) velocity.\(^\text{35} \) When we used \( C_v = 107.2 \text{ mJ/K}, \) \( \kappa = 25 \text{ W/cm K}, \) and \( v = 4 \times 105 \text{ cm/s for GaAs}, \)\(^\text{37} \) the phonon dissipation time was estimated to be \( 4.4 \) ns. This value corresponds quantitatively with our observation of the bunching decay time, and this correspondence supports our interpretation.

In addition, we roughly estimate the number of residual phonons on the basis of the magnitude of \( g^{(2)}_{\text{QD}} \). As seen in Fig. 5(b), the bunching magnitude is almost double. If it is assumed that a phonon cannot exist in the steady state, the phonon scattering rate will double when there is an average of one phonon in the bunching time domain [see Eq. (9)]. In this case, the photon detection probability \( g^{(2)}_{\text{QD}} \) is considered to be doubled. Accordingly, we can roughly estimate that there is one residual phonon in the bunching time domain.

2. Photon emission process of two-exciton system in a CQD

Next, we consider the correlated photon emission between two QDs in a thick barrier CQD. First, we consider the photon detection process in the cross photon correlation experiment. Figure 8 shows the carrier occupation diagram in a CQD, which includes the intraband energy relaxation process in each QD, e.g., \(|1\rangle_g |1\rangle \rightarrow |0\rangle |1\rangle_e \rightarrow |0\rangle |1\rangle_g \rightarrow |0\rangle |0\rangle \). In this figure, the photon detection process of the start signal (QD1) is either \(|1\rangle_0 |0\rangle \rightarrow |0\rangle |0\rangle \) or \(|1\rangle_1 \rightarrow |0\rangle |1\rangle \). The detection processes of the stop signal (QD2) for each start detection process are described as below:

\[
|M_q|^2 \propto |\langle k + q, n_q - 1 | a_q^\dagger | k, n_q \rangle |^2 = n_q \quad \text{: Absorption}
\]

\[
|M_q|^2 \propto |\langle k - q, n_q + 1 | a_q^\dagger | k, n_q \rangle |^2 = n_q + 1 \quad \text{: Emission},
\]

where \( n_q \) is the number of phonons, \( a_q \) and \( a_q^\dagger \) are creation and destruction operators, and \( k \) and \( q \) are exciton and phonon wave numbers. The high scattering rate accelerates the carrier energy relaxation, and accordingly the probability of detecting the stop signal during the phonon survival time is expected to become higher than that in the DT \( \gg 0 \) time domain. On the basis of the above discussion, our observation of the bunching for DT \(< \pm 4 \) ns in Fig. 5(b) is considered to originate from the enhancement in the intraband energy relaxation caused by the residual phonons emitted from the start signal exciton. The decay time of the bunching (\(~\sim 4 \) ns) indicates the phonon dissipation time. Here, let us compare the bunching decay time with the phonon dissipation time estimated using classical statistical mechanics. It is well known that the phonon dissipation time is classically given by

\[
\tau = \frac{3\kappa}{C_v v^2},
\]

where \( C_v \) is the specific heat at a constant volume, \( \kappa \) is the thermal conductivity, and \( v \) is the phonon (sound) velocity.\(^\text{35} \) When we used \( C_v = 107.2 \text{ mJ/K}, \) \( \kappa = 25 \text{ W/cm K}, \) and \( v = 4 \times 105 \text{ cm/s for GaAs}, \)\(^\text{37} \) the phonon dissipation time was estimated to be \( 4.4 \) ns. This value corresponds quantitatively with our observation of the bunching decay time, and this correspondence supports our interpretation.

In addition, we roughly estimate the number of residual phonons on the basis of the magnitude of \( g^{(2)}_{\text{QD}} \). As seen in Fig. 5(b), the bunching magnitude is almost double. If it is assumed that a phonon cannot exist in the steady state, the phonon scattering rate will double when there is an average of one phonon in the bunching time domain [see Eq. (9)]. In this case, the photon detection probability \( g^{(2)}_{\text{QD}} \) is considered to be doubled. Accordingly, we can roughly estimate that there is one residual phonon in the bunching time domain.

The shortest of these detection processes is the (3) pass: QD1: \(|1\rangle_1 |1\rangle \rightarrow |0\rangle |1\rangle \), QD2: \(|0\rangle |1\rangle \rightarrow |0\rangle |0\rangle \). Therefore, in the cross photon correlation measurement, it is considered that the (3) pass is mainly counted and contributes to the \( g^{(2)}_{\text{QD}} \) spectrum. In this detection pass, the QD1 emits a photon at DT \( = 0 \) and then become vacant at DT \( > 0 \). In this case, QD2 must alone emit a photon without an interdot interaction, e.g., \(|0\rangle |1\rangle_e \rightarrow |0\rangle |1\rangle_g \rightarrow |0\rangle |0\rangle \). Here, when we review the discussion in previous sections, the intraband energy relaxation time \textit{without} the interdot interaction is longer than that \textit{with} the interdot interaction. Accordingly, the probability of detecting QD2 is considered to become lower in the immediate aftermath of the QD1 emission: this process results in antibunching. In this case, QD2 will take a long intraband relaxation time before the photon emission and then begin to emit a photon gradually, which results in the long recovery time of the \( g^{(2)}_{\text{QD}} \) antibunching. By the same token, the shortest detection process at DT \( < 0 \) is \(|1\rangle_1 |1\rangle \rightarrow |1\rangle |0\rangle \rightarrow |0\rangle |0\rangle \), which results in the long recovery time of antibunching at DT \( < 0 \) based on the same origin.

At the same time, even if the next carriers are excited in QD1 at DT \( > 0 \), these carriers does not affect the carrier energy relaxation in QD2. This is because the secondarily excited carrier in QD1 is rapidly relaxed energetically by the residual phonon emitted at DT \( = 0 \) and then emits a photon. Therefore, the long energy relaxation time in QD2 is maintained, which keeps the long recovery time of the \( g^{(2)}_{\text{QD}} \) antibunching. In fact, the antibunching recovery times
(4–5 ns) are comparable to the estimated intraband relaxation time described in previous section (~5 ns). As discussed above, the $g^{(2)}_{12}$ antibunching and its long recovery time are attributed to the photon detection processes in the weakly CQD in the presence of the interdot interaction, and these results support our interpretation of PL enhancement outlined in previous sections.

D. Discussion

In addition, we mention other possible ways to interpret our observations. One is the effect of the defect levels, as mentioned in the previous section. In this case, to cause antibunching, the relaxation process through the defect level must be very fast (perhaps less than a few hundred picosecond). This is because the defect level should be filled before the QD1 emission and the filled carriers should disappear immediately after the QD1 emission in order to trap the QD2 carrier. Then, the trapped QD2 carrier will also disappear immediately and the next excited carrier in QD1 or QD2 will fill the defect level again. When the QD1 carrier fills the defect level, the excited carrier in QD2 will relax energetically to the lowest level and emit a photon. In this process, if the energy relaxation time is long, a long recovery time for the antibunching will be observed. However, this kind of fast detection time in the defect level is unlikely, as mentioned in the previous section. Another possibility is the effect of an acoustic phonon as mentioned in this section. The acoustic phonons emitted before the QD1 emission may influence the QD2 emission process. In this case, these residual phonons will accelerate the energy relaxation of the QD2 carriers. This effect would induce the fast recovery time of the $g^{(2)}_{12}$ antibunching, but our observations conflict with this prediction. This is attributed to the fact that the difference in energy of phonons, contributing to the energy relaxation, between QD1 and QD2 causes minimal correlation of photon emitting processes between the two QDs.

Thus, from the above discussions, the acceleration of intraband energy relaxation with the interdot interaction is thought to be the most likely interpretation of our observations.

V. CONCLUSION

We discussed the correlated photon emission and interdot interaction in a thick barrier CQD. By using two-color excitation spectroscopy, we observed that simultaneous carrier creation in two QDs induced an anomalous increment in PL intensity, which indicated the interdot interaction in the CQDs. Based on an analysis of the excitation power dependence of the PL intensity, our proposed interpretation is that an interdot interaction, such as a dipole-dipole interaction, induced an enhancement in the intraband energy relaxation.

We further investigated the photon emission in a CQD using second-order photon correlation spectroscopy. We confirmed that there was a single photon emission from each QD constituting the CQDs. The autocorrelation spectrum depended on the energy relaxation process derived from the exciton-phonon interaction. We revealed that the residual phonons accelerated the intraband energy relaxation resulting in photon bunching. It is noteworthy that the cross photon correlation spectrum between two QDs exhibited a photon antibunching with a long recovery time. We indicated that this observation supported our interpretation that the enhancement in the intraband energy relaxation is caused by the interdot interaction. These results and discussions will provide a significant contribution to the understanding of CQD systems.