Electronic structures in single pair of InAs/GaAs coupled quantum dots with various interdot spacings

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The electronic structures in a single pair of InAs/GaAs coupled quantum dots (CQDs) with various interdot spacings are investigated by performing photoluminescence (PL) and photoluminescence excitation (PLE) measurements. Luminescence from the bonding ($X^+$) and antibonding ($X^-$) states caused by electron-wave-function coupling was observed in the micro-PL spectra of the CQDs. We indicate the contribution of the hole excited states to the PL spectra in QDs based on the results for the spectral dependence on circularly polarized light and the structures of PLE spectra. PLE spectra reveal the electronic structures of the CQD system at higher energy states where both the common excited levels due to the level sharing between the electron excited states and the individual excited levels related to the hole excited states coexist. In addition, we mention that the energy-level mixing due to the strong-wave-function coupling between two QDs influences the decoherence of the carrier relaxation processes. © 2006 American Institute of Physics. [DOI: 10.1063/1.2171809]

INTRODUCTION

The semiconductor quantum dot (QD) has been studied closely because of its attractive physical properties as epitomized by its atomiclike energy levels, which are artificially controllable. Recently, the semiconductor QD has attracted attention as a medium for quantum computing because it has a long carrier coherence time due to the discrete energy levels that originate from three-dimensional quantum confinement. A QD consisting of III-V compound semiconductors is particularly attractive because its energy states can be optically controlled and it is relatively easy to fabricate by the Stranski-Krastanov growth technique. Although Li et al. have already demonstrated a quantum logic gate using the exciton-biexciton correlation in a single GaAs/AlGaAs QD, a single QD has definite limitations in terms of scalability into a large number of quantum bits (qubits). Accordingly, a coupled QD (CQD) system is essential for large numbers of qubits. Although there have been several experimental studies of a single pair of QDs, these reports focused only on the fine structures around the ground exciton states. The coupling mechanism will also differ with the interdot spacing and the energy levels in individual QDs. Therefore, a more detailed understanding of the overall electronic structures, involving higher energy states, in a CQD system with various interdot spacings is important if we are to realize quantum information devices.

In our previous works, we studied the electronic structures and carrier correlation in a single pair of CQDs with narrow and wide interdot spacings, but we have not yet provided detailed discussions of the overall electronic structures in CQDs with various interdot spacings. In this paper, we report the overall electronic structures in a single pair of resonantly coupled InAs/GaAs QD systems with various interdot spacings. We show the emissions that originated from the bonding ($X^+$) and antibonding ($X^-$) states due to the wave-function coupling of an electron and reveal those electronic structures at higher energy states with several interdot spacings. In addition, we mention the influence of the wave-function coupling between two QDs on the coherent carrier relaxation processes.

SAMPLE FABRICATION AND EXPERIMENTAL SETUP

The samples used in this work were self-organized InAs QDs embedded in a GaAs matrix grown by molecular-beam epitaxy (MBE) in a similar way to that described in our previous work. We adopted the indium-flush procedure for the vertical CQD growth in order to avoid any fluctuation in the barrier layer caused by the convexoconcave nature of the first layer QDs when the barrier thickness decreased. The first InAs QD layer was grown on a 200 nm $i$-GaAs buffer layer on a (100) $n$-GaAs substrate, and the indium-flush procedure (635 °C, 100 s) was performed after a 3.5 nm partial GaAs cap layer had been grown over the first QD layer. Then, GaAs barrier layers with various thicknesses and the second InAs QD layer were grown sequentially. We also applied the indium-flush procedure to the second QD layer under the same conditions as those used for the first QD layer, and finally we grew a 100 nm GaAs cap layer. The distances between the first and second InAs wetting layers (WL) were 6, 8, and 10 nm for the three CQDs samples (meaning that the effective thicknesses $d$ of the barrier between the first and second layer QDs were estimated to be 3, 5, and 7 nm). In this study, the thickness of the first InAs layer was kept close to the critical thickness [$1.7\text{ ML (monolayer)}$] of the dot formation in order to achieve a low in-plane QD density. In addition, the indium supplied for the second QD layer was...
limited to a small amount (1.45 ML) in order to fabricate similarly (resonantly) sized upper and lower QDs because the second layer QD growth is accelerated by the strain effect exerted by the first layer QDs.17,18 We also prepared a single QD layer sample using the indium-flush method for reference. Figure 1(a) shows the cross-sectional scanning transmission electron microscope (STEM) image of the CQD samples with d = 7 nm. We clearly confirmed the vertical alignment of two QDs in the image, where we estimated the dot height at ~3 nm and the effective thickness d of the barrier at ~7 nm. We also estimated the dot diameter at ~20 nm and the in-plane density of the QDs at 40–30 pcs/μm² by using a high-resolution scanning electron microscope (HRSEM). In addition, we fabricated an aluminum aperture mask (0.2–0.5 μm φ) on the sample surface by an electron-beam lithography technique to enable us to observe the single CQDs by selecting the aperture hole that showed the luminescence of only one CQD.19 In the macrophotoluminescence (macro-PL) spectra, the luminescence peak from the QD ensemble ranged from 935 to 955 nm. We did not observe any energy splitting caused by the wave-function coupling in the macro-PL spectra because the QD ensemble had a large size distribution due to the thin InAs layer close to the critical thickness of the dot formation.

All the micro-PL (μ-PL) and photoluminescence excitation (μ-PLE) measurements were performed using a microspectroscopy system with a 1 m double monochromator and a cooled charge-coupled device (CCD) detector, in which the spectral resolution was ~20 μeV. The excitation light focusing and the luminescence collection were performed using a microscope objective with a large numerical aperture (spot diameter ~4 μm φ). The samples were cooled to 6 K in a liquid-helium cryostat, and a continuous-wave tunable Ti:sapphire laser was used as the excitation source.

RESULTS AND DISCUSSION

Energy states around ground level

Typical μ-PL spectra from a single CQD are shown in Fig. 1(b), where the excitation energy was 1.41 eV (absorption band of WL) and the excitation intensity was set low enough (<0.1 mW) to prevent multiple excitons from being excited. We also show the PL spectrum of a single QD layer sample as a reference at the top of the figure. As shown in Fig. 1(b), the overall trend of the CQDs is that the spectra have two PL groups on the high- and low-energy sides, unlike the single QD which has only one group. Each PL group consists of two or three peaks that are separated by 2–5 meV, except for the d = 7 nm sample, which is discussed in detail below. The notable feature of the CQDs is that the energy separation between the two PL groups increases with reductions in the barrier thicknesses, as indicated by ΔE in Fig. 1(b). This feature arises from the wave-function coupling of electrons between the upper and lower QDs and these PL groups constitute the bonding (X⁺) and antibonding (X⁻) states, respectively. The barrier thickness dependence of the energy splitting ΔE agrees well with previously reported results.3,14 Of course, ΔE values may include the energy differences originating from the variations of characteristics (size, location, strain, etc.) between two QDs. But, in our observations, many CQDs in the different aperture hole areas show similar PL spectra and ΔE values, although the PL energies varied (1.285–1.350 eV) due to reflection of the inhomogeneity of the QD sizes. As seen in the inset in Fig. 1(b), the small data spread (fudge factor) of ΔE in our samples implies that the upper and lower QDs are of similar (resonantly growing) size in the analogous surrounding conditions. Furthermore, increases in the data spread of ΔE with reductions in the barrier thickness seem to originate from increases in the thickness fluctuations with reductions in the barrier thicknesses because the barrier thickness is a dominant element to affect the wave-function coupling of electrons.14 Other feature of PL spectra is that the higher-energy PL group (X⁻ state) maintains large PL intensity under the weak excitation conditions and has narrow linewidth similar to that of the X⁺ PL group. This indicates that the carrier relaxation from X⁻ to X⁺ state is suppressed. The energy relaxation of carriers under the weak excitation is mainly caused by the phonon emission. As is well known, the carrier relaxation rate induced by the acoustic phonons is ten times less than that by the optical phonons.5 In our cases, the energy splitting ΔE are less than the longitudinal-optical (LO) phonon energy (~35 meV in
InAs QDs): the acoustic phonons dominate the relaxation processes. Therefore, the long relaxation time from $X^{-}$ to $X^{+}$ state due to the contribution of acoustic phonons to the relaxation processes, which is regarded as being longer than the exciton decay time, allows the strong PL intensity for the $X^{+}$ state.

We shall now look more carefully at the multiple PL peaks of each PL group in our QDs. First, we will distinguish the emissions of exciton from those of multiple excitonic states by observing the excitation power dependence of the PL intensities. In the case of multiple excitonic states, the excitation power dependence will be out of linear dependence on the excitation intensity [see the table in Fig. 2].

At low excitation intensities, two peaks with a few meV energy separation dominate the one PL group. When we focus on the two fundamental PL peaks ($X_{1}$ and $X_{2}$) at low excitation intensities, these peaks exhibit a linear dependence on the excitation intensities ($m - 1$). This feature shows that the $X_{1}$ and $X_{2}$ peaks originate from exciton states rather than multiple excitonic states. Much the same is true for the single QD.

The presence of the doublet PL peaks ($X_{1}$ and $X_{2}$) has possibilities of several origins: (1) Charged exciton, (2) different exciton states (involving a dark exciton) caused by the anisotropy of the QD shape, (3) asymmetric transition between an electron in one QD and a hole in the neighboring QD, and (4) excited states of exciton. We shall consider these possibilities in order.

(1) Charged exciton. Many previous studies indicate the existence of a charged exciton in In(Ga)As QDs (Refs. 19–21) and GaAs quantum disks. As is well known, a negatively (positively) charged exciton shows a lower- (higher-) energy PL peak than a PL peak of a neutral exciton. Furthermore, it has been reported that a positively charged exciton in a GaAs quantum disk had the polarization memory to circularly polarized light unlike a neutral exciton which did not depend on circularly polarized light. Here, we show the circular polarization dependences of the PL spectra in our QDs and a single QD in Figs. 3(a)–3(d). As seen in the figures, the $X_{1}$ peak in all samples is strongly dependent on circularly polarized light. The $X_{1}$ peak, therefore, has possibility of a positively charged exciton because of the higher peak energy than the neutral exciton energy ($X_{2}$ peak). But we need to be careful about the origin of extra carriers contributing to the charged exciton formation, which will be supplied from out of QDs or exist preliminarily in QDs. In Ref. 19, it has been reported that the PLE spectra of the positively charged exciton do not have peaks below the WL energy. This suggests that the extra carriers contributing to the charged exciton formation are not excited under the excitation condition of lower energy than the WL energy; this condition leads to create only one pair of an electron and a hole. In other words, in this case, the extra carriers contributing to the charged exciton formation were supplies from out of a QD. In Ref. 21, it has been also reported that the PLE spectra of the positively charged exciton do not have peaks around the WL energy. This suggests that the carriers originating from background doping, which contribute to the charged exciton formation, are compensated by the extra carriers dissociating from excitons in the WL. In other words, in this case, the extra carriers contributing to the charged exciton formation exist preliminarily in a QD. However, in our cases, the structure of $X_{1}$ PLE spectra (see Figs. 4 and 5) has many peaks at the overall excitation energy and we confirmed that the $X_{1}$ peak maintains its intensity at the WL excitation (Figs. 1–3) and the interband excitation (1.55 eV), conflicting with the results of previous works about the charged exciton. These observations suggest that the $X_{1}$ peak does not
originate from extra carriers. Therefore, a charged exciton does not explain our observations.

(2) Different exciton states (involving a dark exciton) due to the QD shape. The doublet exciton states in a QD have been discussed theoretically $^{23-25}$ and have been observed experimentally. $^{26,27}$ This doublet structure with 0.01–1 meV scale energy splitting is considered to be results of an activation of the “bright” exciton mixing with the “dark” exciton through the exchange interaction caused by the anisotropy of the QD shape or magnetic field induced. $^{23-27}$ Furthermore, it has been reported that these doublet peaks depend on linearly polarized light, because of the origin of the QD shape anisotropy. $^{23,26}$ Now, we show the linear polarization dependences of the PL spectra in our $d=3$ nm CQDs in Fig. 3(e). As seen in the figure, the ratios between the $X_1$ and $X_2$ PL intensities are almost same. This result indicates that the $X_1$ and $X_2$ peaks do not relate to the QD shape anisotropy. Therefore, the possibility given here cannot explain our observations. Intrinsically, the splitting energy between the $X_1$ and $X_2$ peaks (1–5 meV) is too large to be explained by the exchange interaction.

(3) Asymmetric transition between an electron in one QD and a hole in the neighboring QD. It has been suggested that an electron of the coupling levels (bonding and antibonding states) in a QD has a possibility to recombine severally with each hole in two QDs because a hole is isolated in each QD due to its large effective mass. $^{14}$ This prediction indicates multiple PL peaks in a PL spectrum of a single QD. In this case, it seems to be roughly expected that the energy separation between two peaks ($X_1$ and $X_2$) will be same in both PL groups ($X^+$ and $X^-$) because the energies of holes in each QD stay constant. But, in our observation, the energy separations between the $X_1$ and $X_2$ peaks are all different in each PL group of every CQDs. Furthermore, the fact that the single QD also has double PL lines conflicts with this prediction, because a single QD would have a single PL line if this prediction was true. In addition, this prediction does not explain the circular polarization memory of the $X_1$ peaks. Therefore, the asymmetric transition cannot explain our observations.

(4) Excited states of exciton. Thus, the possibilities of cases (1)–(3) mentioned above cannot explain our observation. When we focus on the circular polarization memory of the $X_1$ peaks, this feature implies that the $X_1$ states have a nonzero quantum number; it means that the $X_1$ states will be excited states of a neutral exciton. Many hole bound states with a close energy-level separation of a few meV in a QD were expected on the basis of

FIG. 3. [(a)–(d)] Dependence of the PL spectra on circularly polarized light, where the excitation energy is 1.41 eV. The polarization of the excitation light is clockwise and the polarization of the PL detection is both clockwise and anticlockwise. (e) Dependence of the PL spectra on linearly polarized light in $d=3$ nm CQDs, where the excitation energy is 1.41 eV. The polarization angle of the PL detection was varied in 45° increments in a plane vertical to the growth direction, while the polarization angle of the excitation light was fixed.

FIG. 4. PLE spectra in a CQD: (a) $d=3$ nm and (b) $d=5$ nm. The dotted line indicates the coincident peaks in the same PL group (between the $X_1$ and $X_2$ states) and the dashed line indicates the coincident peaks between the two PL groups (the $X^+$ and $X^-$ states).
Electronic structures at higher energy

Next, we discuss the higher excited states of CQDs using the results of PLE measurements. The electronic structures at the higher energy states will inform us about the carrier transfer between two QDs through the electron-wave-function coupling of the excited levels. Moreover, we will also identify the origin of the emission more clearly by means of comparing the peak energy correspondence between PLE spectra. In Figs. 4 and 5, we present the PLE spectra of a single CQD with various barrier thicknesses and a single QD. We can see two general characteristics in the figure. One is that the PLE spectra of the two PL peaks ($X_1$ and $X_2$) in one PL group have many coincident peaks as indicated by the dotted line. Another is that there are several coincident PLE peaks between the two PL groups (between the $X^+$ and $X^-$ states) as indicated by the dashed line, but other peaks are inherent to each group.

The former characteristic, the existence of many coincident PLE peaks in one PL group (between $X_1$ and $X_2$), indicates that these two peaks originate from the same excited level series, which means that the $X_1$ peak is the excited level of the $X_2$ state in a group. In the single QD [Fig. 5(b)], we also observed the same feature, which indicates the pair PL lines ($X_1$ and $X_2$) originating from the same QD. These observations support our suggestion in the previous section that the $X_1$ state is the $p$-like hole excited state. In contrast, in the case of $d=7$ nm CQDs [Fig. 5(a)], the PLE spectra of two PL peaks ($X_1^+$ and $X_2$) are not correspondent, which means that these peaks originate from different QDs in $d=7$ nm CQDs. Thus, comparison of PLE spectra enables us to identify the origin of the emission.

On the other hand, the latter characteristic, the coexistence of both the coincident PLE peaks between the two PL groups and the inherent peaks to each group, indicates the coexistence of two different excited level series in the electronic structure of our CQDs, which are the common excited level series between the $X^+$ and $X^-$ levels and the individual excited level series for each state. In these two excited level series, the individual excited level series is considered to originate from the hole excited states. As mentioned above, many hole bound states with a close energy-level separation of a few meV exist in a QD. Because of a large effective mass of the hole, the hole wave functions will not couple with the neighboring QD. Therefore, the energy levels related to the hole excited states should be observed as individual excited level series for each PL group (each coupling level of $X^+$ and $X^-$). On the contrary, the electron-wave-function coupling between two QDs will induce the common excited levels between the $X^+$ and $X^-$ states. The number of the common excited levels should change with the coupling strength between two QDs; for example, the strong wave-function coupling will result in many common excited levels due to increasing of the number of the electron excited states coupling with the neighboring QD (the level sharing between the $X^+$ and $X^-$ states), which means enhancement of the carrier transfer between two QDs. This prediction can be seen in Figs. 4 and 5. Comparing the PLE spectra of CQDs with various barrier thicknesses, the number of the common

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FIG. 5. PLE spectra in a CQD and single QD: (a) $d=7$ nm CQDs and (b) single QD. Indications of the dotted and the dashed lines are the same as in Fig. 4. A and B indicate the PLE peak examples of the levels which induced the Fano resonance between the $X_{1,2}$ and $X_{xx}$ states.
PLE peaks between the $X^+$ and $X^-$ states increases with reductions of the barrier thicknesses as indicated by the dashed line. This observation directly shows that the level sharing between the $X^+$ and $X^-$ states and the carrier transfer between two QDs are enhanced with reductions of the barrier thickness due to the wave-function coupling of the excited levels.

Thus, we observed the common excited levels due to the level sharing between the electron excited states and the individual excited levels originating from the hole excited states in a CQD system. From these results, we can roughly classify $d=3$ and $5$ nm CQDs into the quantum-mechanical coupling system (strong-coupling region), in which the wave-function coupling and the carrier transfer are large, and $d=7$ nm CQDs into the electromagnetic coupling system (weak-coupling region), in which the wave-function coupling and the carrier transfer are very small and the electromagnetic interaction will be dominant.

Carrier relaxation in CQD system

Additionally, we will mention the carrier relaxation in a CQD system. Gotoh et al. discussed the quantum resonance, namely, the Fano resonance, in a single QD. They suggested that an exciton shows the Fano resonance with a bie exciton because the difference in the relaxation path between the exciton and the biexciton yields the phase difference of the coherent dipole as long as the exciton and the biexciton maintain its coherence. Furthermore they showed its experimental evidences in a single QD in which the PLE spectra of the exciton showed dip structure caused by the existence of the bie exciton. We observed same features in the PLE spectra of our QDs. In Figs. 4 and 5, we also show the PLE spectra of the biexcitonlike states (labeled as $X_{ss}$). With the single QD and $d=7$ nm CQDs, there are dip structures in the $X_{1,2}$ PLE spectra at an energy corresponding to the peaks in the PLE spectra of the $X_{ss}$ states, for example, peaks $A$ and $B$ in the figures (the detail spectra are shown in Fig. 6). This phenomenon suggests that there is Fano resonance between the $X_{1,2}$ and $X_{ss}$ states in our QD as discussed in Ref. 23.

This expression of the quantum resonance will enable us to monitor the coherence of the carrier relaxation in a QD. The strong wave-function coupling will induce many fine energy structures because of the exchange interaction among electrons and holes. Then, the complexity of the relaxation process originating from the mixed energy structures should dissipate the carrier coherence and eliminate the quantum resonance because of the carrier-phonon interaction which is the main rule of the carrier relaxation. This prediction can be seen in our observation. The reductions in the barrier thicknesses lead many weak PLE peaks which blur boundaries between neighboring peaks and yield quasicontinuum band (Figs. 4 and 5). This suggests that the strong wave-function coupling induces many mixing energy states. And in the single QD and $d=7$ nm CQDs which are a noncoupling and the carrier transfer are large, and $d=7$ nm CQDs into the electromagnetic coupling system (weak-coupling region), in which the wave-function coupling and the carrier transfer are very small and the electromagnetic interaction will be dominant.

CONCLUSION

We studied the electronic structures in a single pair of InAs/GaAs CQDs using PL and PLE measurements. The bonding and antibonding states due to the electron-wave-function coupling in the CQD system were confirmed by comparing the $\mu$-PL spectra of several CQDs with different barrier thicknesses. We discussed the origin of the multiple PL peaks and suggested the contribution of the hole excited states to the emissions from the QDs based on the dependence of the emissions on the circularly polarized light and the structures of the PLE spectra. On the basis of the PLE measurements, we showed the electronic structures of a CQDs system in higher energy states. We confirmed the common excited levels due to the level sharing between the electron excited states and the individual excited levels originating from the hole excited states in a CQD system. Moreover, we indicated the influence of wave-function coupling on the decoherence of the carrier relaxation processes in
single CQDs. These results and discussions make a significant contribution to the understanding of CQD systems with a view to realizing quantum information device applications.

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